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(71) Applicant (for all designated States except US): MERCK PATENT GMBH [DE/DE]; Frankfurter Strasse 250, 64293 Darmstadt (DE).

(72) Inventors; and

(75) Inventors/Applicants (for US only): PAN, Junyou [CN/DE]; Sigmund-Freud-Strasse 105, 60435 Frankfurt (DE). MEYER, Frank [DE/DE]; Ladenburger Strasse 46, 69120 Heidelberg (DE).

(74) Common Representative: MERCK PATENT GMBH; Frankfurter Strasse 250, 64293 Darmstadt (DE).

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(54) Title: POLYMER BLENDS AND THEIR USE IN ORGANIC LIGHT EMITTING DEVICES

(57) Abstract: The invention relates to novel polymer blends comprising one or more hole transporting polymers and one or more electron transporting polymers, to the use of these blends in electronic and electrooptical devices, in particular in organic light emitting diodes (OLEDs), and to OLEDs comprising the polymer blends.

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Polymer Blends and their Use in Organic Light Emitting Devices

Field of the Invention

The invention relates to novel polymer blends comprising one or more hole transporting polymers and one or more electron transporting polymers. The invention further relates to the use of these blends in electronic and electrooptical devices, in particular in organic light emitting diodes (OLEDs). The invention further relates to OLEDs comprising the polymer blends.

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Background and Prior Art

OLEDs based on organic polymer materials, also known as polymer light emitting diodes (PLEDs), have drawn much attention due to their potential application in next generation panel displays. Though huge improvement has been achieved in the last years, the performance of the PLEDs, especially the lifetime (particularly for blue PLEDs) still needs further improvement to be commercially successful. Single layer PLEDs, where the hole transport, electron transport and emissive layer are combined into one layer, have the advantage of simple processing, however, they do often show poor lifetime. WO 2004/084260 A2 discloses a PLED wherein the combination of a specific cathode metal with an interlayer between the hole injection layer (HIL) and the light emitting polymer (LEP) is reported to improve the lifetime compared to a conventional single layer PLED.

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An "interlayer" as referred to hereinafter means a layer in an OLED device that is situated either between the hole injection layer (HIL) and the emissive layer (EL), or between the electron injection layer (EIL) and the EL, and is intended to prevent electrons from flowing into the HIL, or holes from flowing into the EIL, respectively. An interlayer for use between the HIL and the EL should usually comprise a material having hole transport and electron blocking property, and an interlayer for use between the EIL and the EL should comprise a material having electron transport and hole blocking property.

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However, the additional interlayer is undesired in mass production. Also,

since its processing is not easy and well controllable, the reliability of the performance of a PLED comprising such an interlayer is usually not sufficient for mass production.

- It is therefore one aim of the present invention to find single layer PLEDs having a lifetime of the interlayer system that is comparable or even better than that of prior art PLEDs. Another aim of the present invention is to provide new materials for use in single layer PLEDs, which have advantageous properties, in particular good processability and high lifetime.
 Another aim the present invention is to extend the pool of PLED materials available to the expert. Other aims of the present invention are immediately evident to the expert from the following detailed description.
- 15 blends as claimed in the present invention. In particular, it was surprisingly found that by using a blend of a hole conducting polymer (hereinafter also referred to as "polymer 1") and an electron conducting polymer (hereinafter also referred to as "polymer 2") as LEP in a single layer PLED device, it is possible to achieve a longer lifetime than in a PLED where polymer 1 has the function of an interlayer and polymer 2 has the function of an emissive layer.
 - WO 2005/053052 A1 discloses a polymer blend comprising a first and a second polymer with triarylamine units, wherein both polymers have hole transport properties. In contrast, the polymer blends according to the present invention are characterized in that the two polymers do transport different types of charge carriers, and preferably either polymer transports only one type of charge carrier.

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Morteani et al., Adv. Mater. 2003, 15(20), 1708 and WO 02/28983 A1 disclose a polymer blend comprising a polymer with fluorene units and benzothiadiazole units and a polymer with fluorene units and triarylamine units, and its use in PLEDs. However, the polymers described in these references are suggested to have a bandgap offset smaller than the exciton binding energy, so that the exciton can be stabilised at the blend interface, thus realising a barrier-free heterojunction, and thus a high efficient green

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PLED using these blends. In contrast, a blend consisting of polymers having a large bandgap offset is usually considered to be advantageous only for use in photovoltaic cells, but disadvantageous for use in PLEDs. This is reported for example by J.J.M. Halls et al., Phys. Rev. B. 1999, 60, pp5721. Also, WO 02/28983 A1 discloses only a narrow, defined range for the molecular weight of the polymers to be used in the blend.

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Birgerson et al., Adv. Mater. 1996, Vol 8, pp982, "Efficient blue-light emitting devices from conjugated polymer blends", discloses a device wherein the emissive layer consists of a blend of PDHPT (poly(2,5-diheptyl-1.4-phenylene-alt-2,5-thienylene)) and PDPP (poly(2,5-diheptyl-2',5'dipentoxybiphenylene)). US 5,378,519 discloses a PLED containing a compound having a skeleton of triarylamin and having a carbonyl group. The compound can be blended with other polymers as emissive layer. Cimrova et al., in Adv. Mater. 1998, Vol 10, pp676, "blue light emitting devices based on novel polymer blends" disclose a device wherein the emitting layer consists of PPBSi (poly(phenylbiphenylylsilylene)), P3V (poly(p-terphenyldiyl-vinylene)) and PBD (2-(4-biphenylyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole)). The two polymers have similar HOMO levels. Berggren et al, Nature 1994, Vol. 372, pp444, discloses PLEDs comprising a polymer blend, wherein the color is varying as a function of the operation voltage. The polymers used in this device are four different thiophene homopolymers.

Cina et al., Proceedings of SPIE Vol. 4279 pp221, discloses a PLED based on a blend of two polyfluorenes prepared by Suzuki cross-coupling, with the first component being an electron transporter and emitter and the second component being a hole transporter. No information on the polymer composition is given. However, it is indicated in the document that emitter and electron transport unit are given by the same unit in the first component. Morgado, et al., describe in Appl. Phys. Lett. 2002, Vol 80, pp2436 a PLED using a blend of homo polymer poly(9,9'-dioctyl-fluorene) (PFO) with an alternating polymer poly(9,9'-dioctylfluorene-alt-benzothiadiazole) (F8BT) as emissive layer, and PPV prepared via standard precursor route as interlayer.
The same blend is dicussed by Wilkinson et al., in Appl. Phys. Lett. 2001, Vol 79 pp171 for the dependence of performance on pixel dimension. Niu et

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al., describe a red PLED consisting of a blend of MEHPPV with terpolymers of dioctyl-fluorene (F8), benzothiadiazole (BT) and dithienylbenzothiadiazole (DBT). The MEHPPV is used as the hole transporting component, polymer backbone of the terpolymers as electron transport unit, and BT and DBT as emitter unit. Here all the HOMOs of electron transporting and emitter units are lower than the hole transporting unit MEHPPV. Suh, et al. report in Adv. Mater. 2003, Vol 15 pp1254, that a blue bispirofluorene polymer can be enhanced by blending it with hole transporting small molecules (HTSM), such as 1,3,5-tris(N,N-bis(4-methoxyphenyl) aminophenyl) benzene (TDAPB), 4,4',4"-tris(N-3-methyl-phenyl-N-phenylamino) tri-phenylamine (MTDATA), N,N'-di(4-(N,N'-diphenyl-amino)phenyl)-N,N'-diphenylbenzidine (DNTPD) and 1,1-bis(4-bis(4-methylphenyl) amino-phenyl)cyclohexane (TAPC). Here, all of the HTMs have HOMOs similar to the blue polymer. Yong Cao et al., study in Science 1999, Vol. 397 pp414 the singlet/triplet ratio in a conjugated polymer attained by blending electron transoport materials with conjugated polymers, OC1C10-PPV and MEHPPV.

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In WO 99/48160 A1, an OLED using a mixture of a hole transporting component (first component), an electron transporting component (second component) and an emissive component (third component) is disclosed, wherein at least one of the first, second and third components forms a type II semiconductor interface, with another of the first, second and third component. The type II interface is defined as an interface in which the minimum energy difference between the highest HOMO and the lowest LUMO state is between levels on different sides of the heterojunction. It is also disclosed that the third component and one of first and second component can be provided as functional moieties, say as pendant group, of the same molecule, say copolymer. However, no further enabling technical disclosure is provided in this direction. In fact, no such blend system with long lifetime has been reported so far.

It was now surprisingly found that by using a blend of two polymers with a large HOMO level offset as claimed in the present invention, a PLED device with improved performance could be achieved, in particular a single layer PLED with comparable or even better lifetime than interlayer PLEDs, thus leading to simple processing and higher reliability in mass production. Also,

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it was surprisingly found that in the blends according to the present invention even polymers with high molecular weight can be used, which could not be expected in view of the prior art.

5 The polymer blends of the present invention can be advantageously used both in PLEDs with and without an interlayer. On the one hand, if the polymer blends according to the present invention are used in a PLED device, it is possible to increase the lifetime of the device without the need of an interlayer, so that the interlayer can be omitted and the device 10 assembly can be simplified. On the other hand, if the polymer blends according to the present invention are used in a PLED device comprising an interlayer, they do still significantly improve the performance of said device.

15 **Summary of the Invention**

The invention relates to a polymer blend comprising

- a first polymer (polymer 1) comprising a unit having hole transporting property (hole transporting unit),
- 20 - a second polymer (polymer 2) comprising a unit having electron transporting property (electron transporting unit) and a unit having exciton formation property (exciton formation unit), and optionally comprising one or more further units having emissive property (emissive unit) and/or exciton formation property, wherein preferably said second 25 polymer comprises at least one unit having emissive property,
 - wherein each of said units has a HOMO ("highest occupied molecular orbital") and a LUMO ("lowest unoccupied molecular orbital"), and the difference between said HOMO and LUMO is hereinafter referred to as the "energy gap",
- 30 wherein the HOMO of both said hole transporting unit of said first polymer and said exciton formation unit of said second polymer are higher, preferably at least 0.2 eV higher, very preferably at least 0.3 eV higher, than the HOMO of said electron transporting unit of said second polymer, with the proviso that a blend that comprises a polymer comprising 35 substituted or unsubstituted fluorene and triarylamine units and further

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comprises a polymer comprising substituted or unsubstituted fluorene and benzothiadiazol units is excluded.

Preferably the LUMO of said exciton formation unit of the second polymer is higher than the LUMO of said electron transporting unit of the second polymer.

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Preferably the HOMO of said hole transporting unit of the first polymer is at least 0,4 eV, very preferably 0.5 eV higher than the HOMO of said electron transporting unit of the second polymer.

Preferably the HOMO of said exciton formation unit of the second polymer is at least 0,4 eV, very preferably 0.5 eV higher than the HOMO of said electron transporting unit of the second polymer.

Optionally the exciton formation unit is itself an emissive unit, i.e. it has exciton formation property and emissive property.

20 Preferably the second polymer contains one or more additional emissive units, wherein the excitation energy is transferred from exciton formation unit to each of said additional emissive units, preferably by Förster transfer, and the energy gap of each of said additional emissive units is smaller than the energy gap of said exciton formation unit.

Preferably the additional emissive units emit blue, green and/or red light.

Preferably the second polymer has a negligible hole mobility compared to the first polymer, i.e. 3 or more than 3 orders of magnitudes lower than the hole mobility of the first polymer.

Preferably the first polymer has a negligible electron mobility compared to the second polymer, i.e. 3 or more than 3 orders of magnitudes lower than the electron mobility of the second polymer.

Preferably the first and second polymer are conjugated polymers.

Optionally, the first polymer comprises one or more further emissive units, which do preferably have a lower HOMO than the HOMO of said hole transporting unit of the first polymer and a higher LUMO than the LUMO of said electron transporting unit of the second polymer, and accept excitation energy from said exciton formation unit of the second polymer by energy transfer, preferably by Förster transfer.

The invention further relates to the use of the polymer blends as described above and below in an electronic or electrooptical device, in particular in polymer light emitting diodes (PLED).

The invention further relates to an electronic or electrooptical device, in particular a PLED, comprising a polymer blend as described above and below.

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Further electronic or electrooptical devices include, without limitation, an organic field effect transistor (OFET), thin film transistor (TFT), organic solar cell (O-SC), organic laser diode (O-laser), organic integrated circuit (O-IC), radio frequency identification (RFID) tag, photodetector, sensor, logic circuit, memory element, capacitor, charge injection layer, Schottky diode, planarising layer, antistatic film, conducting substrate, conducting pattern, photoconductor, electrophotographic element, or organic light emitting transistor (OLET).

25 The invention further relates to an electronic device comprising an anode, a cathode, a semiconductor or emissive layer comprising a polymer blend as described above and below, and an interlayer comprising a material having hole transporting and electron blocking property provided between the anode and the semiconductor or emissive layer. A device with such an 30

interlayer is generally described in WO 2004/084260 A2.

The invention further relates to an electronic device comprising a polymer blend as described above and below, and further comprising a conductive polymer layer and/or a hole transporting layer, wherein the polymer blend is coated directly onto said conductive polymer and/or hole transporting layer.

The invention further relates to an electronic device comprising, in the sequence as described below

- optionally a first substrate,
- an anode layer,
- 5 optionally a hole injection layer,
 - optionally an interlayer comprising a material having electron blocking property,
 - a layer comprising a polymer blend as described above and below,
 - a cathode layer,

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- optionally a second substrate.

The second polymer itself is another object of the invention. Thus, the invention also relates to a polymer comprising an electron transporting unit and an exciton formation unit, and optionally comprising one or more further emissive units and/or exciton formation units, wherein each of said units has a HOMO and a LUMO, and the HOMO of said first exciton formation unit is at least 0.2, preferably at least 0.3 eV higher than the HOMO of said electron transporting unit. Preferably the polymer comprises at least one emissive unit. Further preferably the polymer is a polymer as described for polymer 2 above and below.

Definition of Terms

"Hole transport property" refers to a material or unit capable of transporting holes (i.e. positive charges) injected from a hole injecting material or an anode. "Electron transporting property" refers to a material or unit capable of transporting electrons (i.e. negative charges) injected from an electron injecting material or a cathode. "Exciton formation property" refers to a material or unit, wherein holes and electrons can recombine to form an exciton, or which can form an exciton by optical excitation, i.e by absorbing one photon. "Emissive property" refers to a material or unit which, upon receiving excitonic energy by energy transfer from other units, or by forming an exciton either electrically or optically, undergoes radiative decay to emit light.

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"Electron blocking property" refers to a material or unit which, if coated adjacent to an electron transporting layer in a multilayer structure, prevents the electron flowing through. Usually it has a higher LUMO than the electron transporting material in the adjacent electron transporting layer.

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"Backbone group", unless stated otherwise, means the group/groups that has/have the highest content of all groups present in a polymer, preferably with a ratio $\geq 20\%$, very preferably $\geq 30\%$, in particular $\geq 40\%$, most preferably $\geq 50\%$. Backbone groups can also form electron transporting units, hole transporting units, exciton formation units or emissive units either alone or in combination with other groups. For example, if there are two groups whose contents are clearly higher than those of the other groups present in the polymer, or if there are only two groups present in a polymer, then both groups are considered as backbone groups. Preferably the backbone groups are hole transporting groups or electron transporting groups.

The term "polymer" includes homopolymers and copolymers, e.g. statistical, alternating or block copolymers. In addition, the term "polymer" as used hereinafter does also include dendrimers, which are typically branched macromolecular compounds consisting of a multifunctional core group onto which further branched monomers are added in a regular way giving a tree-like structure, as described for example in M. Fischer and F. Vögtle, *Angew. Chem., Int. Ed.* **1999**, *38*, 885.

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The term "conjugated polymer" means a polymer containing in its backbone (or main chain) mainly C atoms with sp²-hybridisation (or optionally also sphybridisation), which may also be replaced by hetero atoms. In the simplest case this is for example a backbone with alternating C-C single and double (or triple) bonds, but does also include polymers with units like 1,3-phenylene. "Mainly" means in this connection that a polymer with naturally (spontaneously) occurring defects, which may lead to interruption of the conjugation, is still regarded as a conjugated polymer. Also included in this meaning are polymers wherein the backbone comprises for example units like aryl amines, aryl phosphines and/or certain heterocycles (i.e.

conjugation via N-, O-, P- or S-atoms) and/or metal organic complexes (i.e. conjugation via a metal atom).

The term "unit" in a polymer according to the present invention means a repeating unit in a polymer, which may consist of a single monomeric group or may also be formed by two or more monomeric groups. For example, an emissive unit may be formed by an emitter group A^{em} and two polymer backbone groups B and is then given by the formula B-A^{em}-B. Unless stated otherwise, the quantum chemistry simulation (as described above) is carried out on units of this formula.

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The units in polymer 1 and 2 of this invention are preferably of the formula -B-A^x-B-, wherein the groups B denote independently of one another identical or different backbone groups, preferably the same backbone group, and A^x is a group selected from groups having hole transporting property (A^h), electron transporting property (A^e), exciton formation property (A^{ex}), or emissive property (A^{em}). Thus, polymer 1 preferably comprises one or more hole transporting units of formula -B-A^h-B-, wherein A^h is a group having hole transport property. Polymer 2 preferably comprises one or more electron transporting units of formula -B-A^e-B-, wherein A^e is a group having electron transport property, and one or more exciton formation units of formula -B-A^{ex}-B-, wherein A^{ex} is a group having exciton formation property. Furthermore, at least one of polymers 1 and 2, preferably polymer 2, preferably comprises one or more emissive units of formula -B-A^{em}-B-, wherein A^{em} is a group having emissive property.

However, it is also possible that polymer 1 and/or polymer 2 comprise one or more units formed by single groups B, A^h, A^e, A^{ex}, A^{em}, which are not present in form of triads B-A^x-B as described above.

Some important energy levels will be explained hereinafter. For conjugated polymers, important characteristic are the binding energies, which are measured with respect to the vacuum level of the electronic energy levels, especially the "highest occupied molecular orbital" (HOMO), and "lowest unoccupied molecular orbital" (LUMO) levels. These can be measured by photoemission, e.g. XPS (X-ray photoelectron spectroscopy) and UPS

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(ultra-violet photoelectron spectroscopy) or by cyclovoltammetry (hereinafter referred to as CV) for oxidation and reduction. It is well understood in the field that the absolute energy levels are dependent of the method used, and even of the evaluation method for the same method, for example the onset point and peak point on the CV curved give different values. Therefore, a reasonable comparison should be made by the same evaluation method of the same measurement method. More recently, the quantum chemistry method, for example Density Function Theory (hereinafter referred to as DFT), has also become well-established to calculate the molecular orbital, particulary the occupied molecular orbitals; and especially the HOMO levels can be well estimated by this method. Therefore, with the help of DFT, given by commercially available software like for example "Gaussian 03W" (Gaussian, Inc.), the HOMO/LUMO of the different units in the conjugated polymers can be calculated.

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The applicants established a very consistent combination method to determine the energy levels of organic materials. The HOMO/LUMO levels of a set of materials (more than 20 different materials) are measured by CV with a stable evaluation method and also calculated by the DFT of Gaussian 03W with the same correction functional, for example B3PW91 and the same basis set, for example 6-31G(d). The calculated values are then calibrated according to the measured values. Such calibration factor is used for further calculation. As will be shown below, the agreement between calculation and measurement is very good. Therefore, the comparison of the energy levels of this invention is set on a sound base. In doing so, the applicants found that for most of the groups that are equal to or larger than biphenyl, the simulation on the above-mentioned triads, like for example -B-A^h-B-, gives very consistent results.

For determination of energy gaps or bandgaps, the energy levels like the HOMO of different units should be measured or calculated with the same method. The preferred methods used in this invention are calibrated DFT method and CV measurement, most preferably calibrated DFT method, particularly when the concentration of the unit is low in the polymer.

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Unless stated otherwise, the values for the energy gap or bandgap given throughout this invention are obtained by the calibrated DFT method.

It should also be pointed out that the HOMO of the entire conjugated polymer is determined by the highest HOMO of its different units, and the LUMO of the entire conjugated polymer is determined by the lowest LUMO of its different units. Therefore, the HOMO of said second polymer can be determined by the HOMO of its exciton formation unit, and the LUMO of said second polymer can be determined by the LUMO of its electron 10 transport unit.

Unless stated otherwise, groups or indices like Ar¹, R¹, a etc. in case of multiple occurrence are selected independently from each other and may be identical or different from each other. Thus, several different groups might be represented by a single label like "R1".

The term "aryl" or "arylene" means an aromatic hydrocarbon group or a group derived from an aromatic hydrocarbon group. The term "heteroaryl" or "heteroarylene" means an "aryl" or "arylene" group comprising one or more hetero atoms. The terms "alkyl", "aryl", "heteroaryl" etc. also include multivalent species, for example alkylene, arylene, "heteroarylene" etc.

The term "carbyl group" as used above and below denotes any monovalent or multivalent organic radical moiety which comprises at least one carbon atom either without any non-carbon atoms (like for example -C=C-), or optionally combined with at least one non-carbon atom such as N, O, S, P, Si, Se, As, Te or Ge (for example carbonyl etc.). The terms "hydrocarbon group", and "hydrocarbyl group" denote a carbyl group that does additionally contain one or more H atoms and optionally contains one or more hetero atoms like for example N, O, S, P, Si, Se, As, Te or Ge.

A carbyl or hydrocarbyl group comprising a chain of 3 or more C atoms may be linear, branched and/or cyclic, including spiro and/or fused rings.

35 Preferred carbyl and hydrocarbyl groups include alkyl, alkoxy, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy and alkoxycarbonyloxy, each of which is

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optionally substituted and has 1 to 40, preferably 1 to 25, very preferably 1 to 18 C atoms, furthermore optionally substituted aryl or aryloxy having 6 to 40, preferably 6 to 25 C atoms, furthermore alkylaryl, arylalkyl, alkylaryloxy, arylalkyloxy arylcarbonyl, aryloxycarbonyl, arylcarbonyloxy and aryloxycarbonyloxy, each of which is optionally substituted and has 6 to 40, preferably 6 to 25 C atoms.

The carbyl or hydrocarbyl group may be a saturated or unsaturated acyclic group, or a saturated or unsaturated cyclic group. Unsaturated acyclic or cyclic groups are preferred, especially alkenyl and alkinyl groups (especially ethinyl). Where the C₁-C₄₀ carbyl or hydrocarbyl group is acyclic, the group may be linear or branched.

The C₁-C₄₀ carbyl or hydrocarbyl group includes for example: C₁-C₄₀ alkyl, C₂-C₄₀ alkenyl, C₂-C₄₀ alkinyl, C₃-C₄₀ allyl group, C₄-C₄₀ alkyldienyl, C₄-C₄₀ polyenyl, C₆-C₄₀ aryl, C₆-C₄₀ aryloxy, C₆-C₄₀ alkylaryl, C₆-C₄₀ arylalkyl, C₆-C₄₀ alkylaryloxy, C₆-C₄₀ arylalkyloxy, C₆-C₄₀ heteroaryl, C₄-C₄₀ cycloalkyl, C₄-C₄₀ cycloalkenyl, and the like. Very preferred are C₁-C₂₀ alkyl, C₂-C₂₀ alkenyl, C₂-C₂₀ alkinyl, C₃-C₂₀ allyl, C₄-C₂₀ alkyldienyl, C₆-C₁₂ aryl, C₆-C₂₀ arylalkyl and C₆-C₂₀ heteroaryl.

Further preferred carbyl and hydrocarbyl groups include straight-chain, branched or cyclic alkyl with 1 to 40, preferably 1 to 25 C-atoms, which is unsubstituted, mono- or polysubstituted by F, Cl, Br, I or CN, and wherein one or more non-adjacent CH₂ groups are optionally replaced, in each case independently from one another, by -O-, -S-, -NH-, -NR⁰-, -SiR⁰R⁰⁰-, -CO-, -COO-, -OCO-, -O-CO-O-, -S-CO-, -CO-S-, -CO-NR⁰-, -NR⁰-CO-, -NR⁰-CO-NR⁰⁰-, -CY¹=CY²- or -C=C- in such a manner that O and/or S atoms are not linked directly to one another, wherein Y¹ and Y² are independently of each other H, F, Cl or CN, and R⁰ and R⁰⁰ are independently of each other H or an optionally substituted aliphatic or aromatic hydrocarbon with 1 to 20 C atoms.

R⁰ and R⁰⁰ are preferably selected from H, straight-chain or branched alkyl with 1 to 12 C atoms or aryl with 6 to 12 C atoms.

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Halogen is F, Cl, Br or I.

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Preferred alkyl groups include, without limitation, methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, 2-methylbutyl, n-pentyl, s-pentyl, cyclopentyl, n-hexyl, cyclohexyl, 2-ethylhexyl, n-heptyl, cycloheptyl, n-octyl, cyclooctyl, dodecanyl, trifluoromethyl, perfluoro-n-butyl, 2,2,2-trifluoroethyl, perfluorooctyl, perfluorohexyl etc.

Preferred alkenyl groups include, without limitation, ethenyl, propenyl, butenyl, pentenyl, cyclopentenyl, hexenyl, cyclohexenyl, heptenyl, cycloheptenyl, octenyl, cyclooctenyl etc.

Preferred alkinyl groups include, without limitation, ethinyl, propinyl, butinyl, pentinyl, hexinyl, octinyl etc.

Preferred alkoxy groups include, without limitation, methoxy, ethoxy, 2-methoxyethoxy, n-propoxy, i-propoxy, n-butoxy, i-butoxy, s-butoxy, t-butoxy, 2-methylbutoxy, n-pentoxy, n-hexoxy, n-heptoxy, n-octoxy etc.

Preferred amino groups include, without limitation, dimethylamino, methylamino, methylamino, phenylamino, etc.

Aryl groups may be mononuclear, i.e. having only one aromatic ring (like for example phenyl or phenylene), or polynuclear, i.e. having two or more aromatic rings which may be fused (like for example napthyl or naphthylene), individually covalently linked (like for example biphenyl), and/or a combination of both fused and individually linked aromatic rings. Preferably the aryl group is an aromatic group which is substantially conjugated over substantially the whole group.

Preferred aryl groups include, without limitation, benzene, biphenylene, triphenylene, [1,1':3',1"]terphenyl-2'-ylene, naphthalene, anthracene, binaphthylene, phenanthrene, pyrene, dihydropyrene, chrysene, perylene, tetracene, pentacene, benzpyrene, fluorene, indene, indenofluorene, spirobifluorene, etc.

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Preferred heteroaryl groups include, without limitation, 5-membered rings like pyrrole, pyrazole, imidazole, 1,2,3-triazole, 1,2,4-triazole, tetrazole, furan, thiophene, selenophene, oxazole, isoxazole, 1,2-thiazole, 1,3thiazole, 1,2,3-oxadiazole, 1,2,4-oxadiazole, 1,2,5-oxadiazole, 1,3,4oxadiazole, 1,2,3-thiadiazole, 1,2,4-thiadiazole, 1,2,5-thiadiazole, 1,3,4thiadiazole, 6-membered rings like pyridine, pyridazine, pyrimidine, pyrazine, 1,3,5-triazine, 1,2,4-triazine, 1,2,3-triazine, 1,2,4,5-tetrazine, 1,2,3,4-tetrazine, 1,2,3,5-tetrazine, and fused systems like carbazole, indole, isoindole, indolizine, indazole, benzimidazole, benzotriazole, purine, naphthimidazole, phenanthrimidazole, pyridimidazole, pyrazinimidazole, quinoxalinimidazole, benzoxazole, naphthoxazole, anthroxazole, phenanthroxazole, isoxazole, benzothiazole, benzofuran, isobenzofuran, dibenzofuran, quinoline, isoquinoline, pteridine, benzo-5,6-quinoline, benzo-6,7-quinoline, benzo-7,8-quinoline, benzoisoquinoline, acridine. phenothiazine, phenoxazine, benzopyridazine, benzopyrimidine, quinoxaline, phenazine, naphthyridine, azacarbazole, benzocarboline, phenanthridine, phenanthroline, thieno[2,3b]thiophene, thieno[3,2b]thiophene, dithienothiophene, dithienopyridine, isobenzothiophene, dibenzothiophene, benzothiadiazothiophene, or combinations thereof. The heteroaryl groups may be substituted with alkyl. alkoxy, thioalkyl, fluoro, fluoroalkyl or further aryl or heteroaryl substituents.

Preferred arylalkył groups include, without limitation, 2-tolyl, 3-tolyl, 4-tolyl, 2,6-dimethylphenyl, 2,6-diethylphenyl, 2,6-di-i-propylphenyl, 2,6-di-t-butylphenyl, o-t-butylphenyl, m-t-butylphenyl, p-t-butylphenyl, 4-phenoxyphenyl, 4-fluorophenyl, 3-carbomethoxyphenyl, 4-carbomethoxyphenyl etc.

Preferred alkylaryl groups include, without limitation, benzyl, ethylphenyl, 2-30 phenoxyethyl, propylphenyl, diphenylmethyl, triphenylmethyl or naphthalinylmethyl.

Preferred aryloxy groups include, without limitation, phenoxy, naphthoxy, 4-phenylphenoxy, 4-methylphenoxy, biphenyloxy, anthracenyloxy, phenanthrenyloxy etc.

The aryl, heteroaryl, carbyl and hydrocarbyl groups optionally comprise one or more subtituents, preferably selected from silyl, sulpho, sulphonyl, formyl, amino, imino, nitrilo, mercapto, cyano, nitro, halogen, C₁₋₁₂alkyl, C₆₋₁₂ aryl, C₁₋₁₂ alkoxy, hydroxy and/or combinations thereof. The optional substituents may comprise all chemically possible combinations in the same group and/or a plurality (preferably two) of the aforementioned groups (for example amino and sulphonyl if directly attached to each other represent a sulphamoyl radical).

- 10 Preferred substituents include, without limitation, solubilising groups such as alkyl or alkoxy, electron withdrawing groups such as fluorine, nitro or cyano, and substituents for increasing glass transition temperature of the polymer such as bulky groups, e.g. t.-butyl or optionally substituted aryl.
- Preferred substituents include, without limitation, F, Cl, Br, I, -CN, -NO₂, -NCO, -NCS, -OCN, -SCN, -C(=O)NR⁰R⁰⁰, -C(=O)X⁰, -C(=O)R⁰, -NR⁰R⁰⁰, optionally substituted silyl, aryl or heteroaryl with 4 to 40, preferably 6 to 20 C atoms, and straight chain or branched alkyl, alkoxy, alkylcarbonyl, alkoxy-carbonyl, alkylcarbonlyoxy or alkoxycarbonyloxy with 1 to 20, preferably 1 to 12 C atoms, wherein one or more H atoms are optionally replaced by F or Cl, wherein R⁰ and R⁰⁰ are as defined above and X⁰ is halogen.

Detailed Description of the Invention

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By using a polymer blend of the present invention comprising a hole conducting polymer (polymer 1) and an electron conducting polymer (polymer 2) as LEP in a single layer PLED device, it is possible to achieve a lifetime that is longer than in a multilayer PLED device using polymer 1 as interlayer and polymer 2 as LEP.

In a PLED device according to the present invention it is desired, and therefore preferred, to keep the exciton density as low as possible on the backbones of the polymers forming the blend, i.e. the electron transporting backbone in case of polymer 2, and hole transporting backbone in case of polymer 1. In other words, it is preferred to maintain the polymer backbones in a single carrier state, and to keep the excitons mainly only on exciton

formation or emissive units. This is achieved by using a blend system containing polymer 1 and polymer 2, wherein both the hole transporting unit of said polymer 1 and the exciton formation unit of said polymer 2 have HOMOs that are preferably at least 0.3eV, very preferably 0,4eV, most preferably 0.5eV higher than the electron transporting unit of polymer 2; and polymer 2 contains at least one exciton formation unit, preferably whose HOMO is higher than the HOMO of the backbone unit of polymer 1, and a LUMO that is higher than the backbone unit of polymer 2. The exciton formation unit is itself an emissive unit if a) there is no energy transfer to further emissive unit, or b) no further emissive unit is present either in polymer 1 or polymer 2, or c) the energy transfer to the further emissive units is only partial.

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The ratio of the hole transporting unit in polymer 1 is preferably from 25 to 99 mol%.

The ratio of the electron transporting unit in polymer 2 is preferably from 25 to 99 mol%.

- The ratio of the excition formation unit in polymer 2 is preferably from 0.01 to 10 mol%, very preferably from 0.01 to 4 mol %, most preferably from 0.01 to 2.5 mol %. For some uses a polymer 2 wherein the ratio of the exciton formation unit is at least 0.1 mol % is preferred.
- The ratio of the further emissive units in polymer 2 and/or polymer 1 is preferably from 0.01 to 10 mol %, very preferably from 0.01 to 4 mol %; most preferably from 0.01 to 2.5 mol %. For some uses a polymer 2 wherein the ratio of the further emissive units is at least 0.1 mol % is preferred.

The proportion of polymer 1 in the blend is preferably from 0.1 to 50 mol %, very preferably from 5 to 25 mol %, most preferably from 10 to 20 mol %.

The proportion of polymer 2 in the blend is preferably from 50 to 99.9 mol%, very preferably from 75 to 95 mol %, most preferably from 80 to 90 mol %.

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Especially preferred is a blend consisting exclusively of polymer 1 and polymer 2.

- The weight average molecular weight M_W of polymer 1 in the blend is preferably from 10,000 to 900,000 , very preferably from 50,000 to 500,000, most preferably from 200,000 to 400,000.
- The weight average molecular weight M_W of polymer 2 in the blend is preferably from 10,000 to 900,000 , very preferably from 50,000 to 500,000, most preferably from 200,000 to 400,000.

In addition to polymers 1 and 2, the polymer blend may comprise one or more further polymers, preferably selected from polymers having one or more of electron conducting, hole conducting, exciton formation and emissive property.

Preferably polymer 1 and 2 comprise one or more groups A^x and B as defined above selected from phenylene, biphenylene, napthalene, anthracene, phenanthrene, dihydrphenanthrene, fluorene, bifluorene, spirobifluorene, phenylene-vinylene, carbazole, pyrene, perylene, 9,10-dihydrophenan-threne, fused thiophene like thieno[2,3b]thiophene or thieno[3,2b] thiophene, dithienothiophene, dibenzothiphene, phenanthroline, trans-indenofluorene, cis-indenonfluorene, debenzolindenofluorene, indenonaphtharene, triarylamine, or derivatives thereof.

Preferred backbone groups B are those of formula I (cis- or transindenofluorene derivatives)

$$(Ar^{1})_{a}$$

$$(Ar^{2})_{b}$$

$$(Ar^{2})_{b}$$

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- 19 -

wherein

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are independently of each other, and in case of multiple A, B and B' occurrence independently of one another, a divalent group, preferably selected from -CR¹R²-, -NR¹-, -PR¹-, -O-, -S-, -SO-, -SO₂-, -CO-, -CS-, -CSe-, -P(=O)R¹-, -P(=S)R¹- and -SiR¹R²-.

R¹ and R² are independently of each other identical or different groups selected from H, halogen, -CN, -NC, -NCO, -NCS, -OCN, -SCN, -C(=O)NR⁰R⁰⁰, -C(=O)X, -C(=O)R⁰, -NH₂, -NR⁰R⁰⁰, -SH, -SR⁰, -SO₃H, -SO₂R⁰, -OH, -NO₂, -CF₃, -SF₅, optionally substituted silyl, or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and optionally the groups R¹ and R² form a spiro group with the fluorene moiety to which they are attached,

Χ is halogen,

R⁰ and R⁰⁰ 20 are independently of each other H or an optionally substituted carbyl or hydrocarbyl group optionally comprising one or more hetero atoms,

each g is independently 0 or 1 and each corresponding h in the same subunit is the other of 0 or 1,

is an integer ≥ 1, m

Ar¹ and Ar² are independently of each other mono- or polynuclear aryl or 30 heteroraryl that is optionally substituted and optionally fused to the 7,8-positions or 8,9-positions of the indenofluorene group,

a and b are independently of each other 0 or 1,

If the groups R¹ and R² form a spiro group with the fluorene group to which 35 they are attached, it is preferably spirobifluorene.

In formula I the term 'subunit' means the group

Accordingly, a subunit in formula I wherein g is 1 and h is 0 is of structure Ia, and a subunit wherein g is 0 and h is 1 is of structure Ib:

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The groups of formula I are preferably selected from the following subformulae:

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$$R^{1} R^{1}$$

$$R^{1} R^{1}$$

$$R^{1} R^{1}$$

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$$\begin{array}{c}
 & (R)_{r} \\
 & (R)_{r}
\end{array}$$

wherein R¹ is as defined in formula I, r is 0, 1, 2, 3 or 4, and R has one of the meanings of R¹.

R is preferably F, Cl, Br, I, -CN, -NO₂, -NCO, -NCS, -OCN, -SCN,
C(=O)NR⁰R⁰⁰, -C(=O)X⁰, -C(=O)R⁰, -NR⁰R⁰⁰, optionally substituted silyl, aryl or heteroaryl with 4 to 40, preferably 6 to 20 C atoms, or straight chain, branched or cyclic alkyl, alkoxy, alkylcarbonyl, alkoxycarbonyl, alkylcarbonlyoxy or alkoxycarbonyloxy with 1 to 20, preferably 1 to 12 C atoms, wherein one or more H atoms are optionally replaced by F or Cl, and wherein R⁰, R⁰⁰ and X⁰ are as defined above.

Preferred groups of formula I are selected from the following subformulae:

wherein

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- is H, halogen or optionally fluorinated, linear or branched alkyl or alkoxy with 1 to 12 C atoms, and is preferably H, F, methyl, i-propyl, t-butyl, n-pentoxy, or trifluoromethyl, and
- 30 L' is optionally fluorinated, linear or branched alkyl or alkoxy with 1 to 12 C atoms, and is preferably n-octyl or n-octyloxy.

The groups of formula I are also suitable as electron transporting groups A^e when used with other groups with low LUMO, especially preferably those of formula I1, very preferably those of formula I1b.

The total ratio of groups of formula I in polymer 1 is preferably from 10 to 80, very preferably from 30 to 70, most preferably from 40 to 60 mol%.

The total ratio of groups of formula I in polymer 2 is preferably from 1 to 95 %, very preferably from 5 to 60 % and most preferably from 10 to 50 mol%.

Preferred hole transporting groups A^h are those of formula II (triarylamine derivatives):

10 Ar^{3} | $-[-Ar^{1}-Y-Ar^{2}]_{m}$

wherein

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Y is N, P, P=O, PF₂, P=S, As, As=O, As=S, Sb, Sb=O or Sb=S, preferably N,

Ar¹ which may be the same or different, denote, independently if in different repeat units, a single bond or an optionally substituted mononuclear or polynuclear aryl group,

Ar² which may be the same or different, denote, independently if in different repeat units, an optionally substituted mononuclear or polynuclear aryl group,

Ar³ which may be the same or different, denote, independently if in different repeat units, an optionally substituted mononuclear or polynuclear aryl group, which may be optionally substituted by a bridging group linking different chain residues of formula II, and

m is 1, 2 or 3.

The groups of formula II are preferably selected from the following subformulae:

$$\begin{array}{c|c}
(R)_s \\
(R)_r \\
N \end{array}$$
|11

$$\begin{array}{c|c}
(R)_s & (R)_r \\
\hline
(R)_r & (R)_r
\end{array}$$
| 10

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$$(R)_{s}$$

$$(R)_{r}$$

wherein R and r are as defined above and s is 0, 1, 2, 3, 4 or 5.

Particularly preferred groups of formula II are selected from the following subformulae:

wherein L is as defined above.

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Very preferred hole transporting groups A^h are those of formula II1. 15

> The groups of formula II are also suitable as exciton formation groups A^{ex}, especially preferably those of formula II2.

20 The total ratio of groups of formula II in polymer 1 is preferably from 10 to 80, very preferably from 30 to 70, most preferably from 40 to 60 mol%.

The total ratio of groups of formula II in polymer 2 is preferably from 0.01 to 10 %, very preferably from 0.1 to 5 % and most preferably from 1 to 2.5 mol%.

Polymer 2 may comprise one or more groups of formula I and one or more groups of formula II as exciton formation unit, or as part of an exciton formation unit, in a concentration so that they act as hole trap instead of hole transport material. Typical concentrations are from 0.01 to 10 %, very preferably from 0.1 to 5 % and most preferably from 1 to 2.5 mol%.

Further preferred hole transporting groups A^h are those of formula III (thiophene derivatives):

$$-(T^{1})_{c}-(Ar^{4})_{d}-(T^{2})_{e}-(Ar^{5})_{f}-$$
 III

wherein

T¹ and T²

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are independently of each other selected from thiophene, selenophene, thieno[2,3b]thiophene, thieno[3,2b]thiophene, dithienothiophene and pyrrole, all of which are optionally substituted with one or more identical or different groups R⁵,

Ar⁴ and Ar⁵

are independently of each other mononuclear or polynuclear aryl or heteroaryl, which is optionally substituted with one or more identical or different groups R⁵, and is optionally fused to the 2,3-positions of one or both of the adjacent thiophene or selenophene groups,

15 R⁵

has in case of multiple occurrence independently of one another one of the meanings of R¹ in formula I that is different from H, and optionally two or more groups R⁵ form a fused ring group with one or more of the groups T^{1,2} and Ar^{4,5}, wherein said fused ring group is fully or partially unsaturated and optionally substituted,

20

are independently of each other 0, 1, 2, 3 or 4, with $1 < c+e \le 6$,

25 d and f

c and e

are independently of each other 0, 1, 2, 3 or 4.

The groups T¹ and T² are preferably selected of

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$$R^{5}$$
 R^{5}

thiophene-2,5-diyl,

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selenophene-2,5-diyl,

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thieno[3,2b]thiophene-2,5-diyl,

thieno[2,3b]thiophene-2,5-diyl,

dithienothiophene-2,6-diyl or

pyrrole-2,5-diyl,

wherein R⁰ is as defined in formula I and R⁵ is as defined in formula III.

The groups of formula III are preferably selected from the following subformulae:

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III2

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$$R'$$
 R'
 R'
 R'
 R'
 R'
 R'
 R'

- 31 -

wherein Y' is CH or N, X' is S or Se, R⁰, R and r are as defined above, and R', R", R" and R"" have independently of each other one of the meanings of R.

Particularly preferred groups of formula III are selected from the following subformulae:

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wherein the thiophene, thienothiophene and phenyl groups may also be substituted with one or more groups R' as defined above, R^0 is as defined above and is preferably C_{1-8} -alkyl, very preferably methyl.

The ratio of units of formula III in polymer 1 is preferably from 10 to 80, very preferably from 30 to 70, most preferably from 40 to 60 mol%.

- In addition to the groups of formula I, II and/or III as disclosed above and below, polymer 1 may comprise one or more further groups preferably selected from groups have hole transporting properties. Suitable hole transporting groups A^h include, without limitation benzidine, triarylphosphine, phenothiazine, phenoxazine, dihydrophenazine, thianthrene, dibenzo-p-dioxine, phenoxathiine, carbazole, azulene, pyrrole and furan derivatives, or further O-, S- or N-containing heterocycles preferably having a high HOMO. The amount of said additional groups in the polymer is preferably from 1 to 15 mol%.
- In addition, polymer 2 may comprise one or more units of formula I and one or more units of formula II as exciton formation unit in a concentration so that they act as hole trap instead of hole transport material. Typical concentrations are from 0.01 to 15, very preferably from 0.1 to 10, most preferably from 1 to 5 mol%.

Further preferred backbone groups B are those of formula IV (phenanthrene derivatives), as disclosed for example in WO 2005/104264 A1:

$$\begin{array}{c|c}
R^{1} & R^{2} \\
\hline
-\left[Ar^{6} X^{1}\right]_{g} & -\left[X^{2}\right]_{g} Ar^{7} \\
\hline
\end{array}$$

30 wherein

- R¹ and R² have independently of each other one of the meanings given in formula I.
- 35 X¹ and X² are independently of each other -CR¹=CR¹-, -C=C- or -N-Ar⁸-

- Ar⁶⁻⁸ are in case of multiple occurrence independently of one another a bivalent aromatic or heteroaromatic ring system having from 2 to 40 C atoms, which is optionally substituted by one or more groups R¹ as defined in formula I,
- g is in each occurrence independently of one another 0 or 1,
- h is in each occurrence independently of one another 0, 1 or 2.
- The groups of formula IV are preferably selected from the following subformulae:

- wherein R' and R" are as defined above, and are preferably alkyl or alkoxy with 1 to 12 C atoms or aryl or heteroaryl with 5 to 12 C atoms that is optionally substituted.
 - The ratio of the groups of formula IV in polymer 2 is preferably from 1 to 95 %, very preferably from 5 to 60 %, and most preferably from 10 to 50 mol%.
- Further preferred backbone groups B are those of formula V (dihydrophenanthrene derivatives) as disclosed for example in WO 2005/014689 A2:

$$-\left[-Ar^{6}\left[-X^{\frac{1}{3}}\right]_{h}\right]^{\frac{R^{1}}{2}} - \left[-X^{\frac{2}{3}}\right]_{g}Ar^{\frac{7}{3}} - V$$

wherein R^1 and R^2 are as defined in formula I, R^3 and R^4 have independently of each other one of the meanings of R^1 , and $Ar^{6,7}$, $X^{1,2}$, g and h are as defined in formula IV.

The groups of formula V are preferably selected from the following subformulae.

wherein R¹⁻⁴ are as defined above.

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The total ratio of groups of formula V in polymer 2 is preferably from 1 to 90 %, very preferably from 5 to 60 %, and most preferably from 10 to 50 mol%.

The groups of formula V2 are also suitable as emissive groups A^{em}.

Further preferred backbone groups B are those of formula VI (spirobifluorene derivatives), as disclosed for example in WO 03/020790 A1:

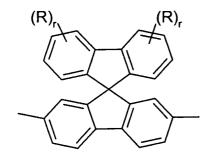
wherein Ar^{6,7}, X^{1,2}, g and h are as defined in formula IV,

- X is in each occurrence independently of one another CH, CR¹ or N,
- is in each occurrence independently of one another a single bond, CR^5R^6 , CR^5R^6 - CR^5R^6 , $CR^5=CR^6$, $CR^6=CR^6$
 - R^{1,2} have independently of each other the meanings given in formula I,
- 20 R^{5,6} have independently of each other one of the meanings of for R¹,
 - m is in each occurrence independently of one another 0, 1, 2, or 3, preferably 0, 1, or 2, very preferably 0 or 1,
- n is in each occurrence independently of one another 0, 1, 2, 3, or 4, preferably 0, 1, or 2, very preferably 1 or 2.

The groups of formula VI are preferably selected from the following subformulae:

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VI1

5 (R)_r (R)_r VI2

wherein R and r are as defined above, and Ar has one of the meanings of Ar⁶ and is preferably 1,4-phenylene that is optionally substituted by one or more groups L as defined above.

Particularly preferred groups of formula VI are selected from the following subformulae:

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wherein r and L are as defined above, and L" is H or linear, branched or cyclic alkyl or alkoxy having 1 to 20 C atoms, or optionally substituted aryl having 5 to 30 C atoms, or -N(Ar)₂, wherein Ar has one of the meanings of Ar⁶ as defined above.

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The groups of formula VI1a wherein L" is -N(Ar)2 are also suitable as hole

transporting groups Ah.

The groups of formula VI2 and VI2a are also suitable as emissive groups Aem.

5 The total ratio of groups of formula VI in polymer 2 is preferably from 1 to 95 %, very preferably from 5 to 60 % and most preferably from 10 to 50 mol%.

Further preferred backbone groups B are those of formula VII (fluorene derivatives):

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$$\begin{array}{c|c}
 & R^{1} \\
\hline
 & Ar^{6} - [X^{1}]_{g} \\
\hline
 & X \\
\hline
 & X \\
\hline
 & X \\
\hline
 & X \\
\hline
 & X^{2} \\
\hline
 & (R^{2})_{m}
\end{array}$$
VII

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wherein R^{1,2} have in each occurrence independently of one another one of the meanings given in formula I, and Ar^{6,7}, X^{1,2}, g and h are as defined in formula IV.

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The groups of formula VII are preferably selected from the following subformulae, as disclosed for example in US 5,962,631:

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wherein R and r are as defined above, and Ar has one of the meanings of Ar⁶ and is preferably 1,4-phenylene that is optionally substituted by one or more groups L as defined above.

Particularly preferred groups of formula VII are selected from the following subformulae:

5 L" L" VII1a

10 L" L" VIII2a

- wherein r and L are as defined above, and L" is H or linear, branched or cyclic alkyl or alkoxy having 1 to 20 C atoms, or optionally substituted aryl having 5 to 30 C atoms, or -N(Ar)₂, wherein Ar has one of the meanings of Ar⁶ as defined above.
- The groups of formula VII2 and VII2a are also suitable as emissive groups A^{em}.

Further preferred backbone groups B are those selected from the following formulae:

25 - Binaphthyl groups of formula VIII, as disclosed for example in WO 2006/063852 A1:

wherein R, Ar⁶, Ar⁷, X¹, X², r, g and h are as defined above, and t is 0 or 1.

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 Groups of formula IX, as disclosed for example in WO 2005/056633A1, EP1344788A1 and WO2007/043495A1:



Preferred groups of formula IX are selected from the following subformulae:

wherein R¹ and R² are as defined above,

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- Groups of the following formulae, as disclosed for example in WO 2005/033174 A1:

$$\begin{array}{c|c}
--Ar^8 ---Ar^9 -\\
 & \downarrow \\
 & X^3 --- X^4
\end{array}$$
XI

wherein

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Ar⁸ and Ar⁹ independently of each other denote a trivalent aromatic hydrocarbon group or a trivalent heterocyclic group,

X³ and X⁴ independently of each other denote O, S, C(=O), S(=O),

$$SO_2$$
, $C(R^1)(R^2)$, $Si(R^1)(R^2)$, $N(R^1)$, $B(R^1)$, $P(R^1)$ or $P(=O)(R^1)$,

 X^5 and X^6 independently of each other denote N, B, P, C(R¹) or Si(R¹),

R^{1,2} are as defined above,

and wherein

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10 X³ and Ar9 are bonded to adjacent carbon atoms in the aromatic ring of Ar9,

X⁴ and Ar⁸ are bonded to adjacent carbon atoms in the aromatic ring of Ar⁹,

X⁵ and Ar⁹ are bonded to adjacent atoms in the aromatic ring of Ar⁸, and

X⁶ and Ar⁸ are bonded to adjacent atoms in the aromatic ring of Ar⁹.

The groups of formula X are preferably of the following subformula:

- Groups of formula XII, as disclosed for example in WO2003/099901A1:

$$\begin{array}{c|c}
X^{7} & X^{8} \\
\hline
-Ar' & Ar'' & (Z)_{z}
\end{array}$$
XII

wherein

35 Ar' and Ar" denote an aromatic hydrocarbon group or heterocyclic group;

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one of X^7 and X^8 denotes C(=0) or $C(R^1)(R^2)$ and the other denotes O, S, C(=O), S(=O), SO₂, $C(R^1)(R^2)$, $Si(R^1)(R^2)$, $N(R^1)$, $B(R^{1}), P(R^{1}) \text{ or } P(=0)(R^{1}),$

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Q is
$$X^9$$
, X^9 - X^{10} or X^{11} = X^{12} ,

X⁹ and X¹⁰ denote independently of each other O, S, C(=O), S(=O), SO_2 , $C(R^1)(R^2)$, $Si(R^1)(R^2)$, $N(R^1)$, $B(R^1)$, $P(R^1)$ or $P(=0)(R^{1})$,

 X^{11} and X^{12} denote independently of each other N, B, P, C(R1) or $Si(R^1)$,

denotes -CR¹=CR²- or -C≡C-. 15 Ζ

> is 0 or 1, Z

R^{1,2} are as defined above.

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The groups of formula XII are preferably selected from the following subformulae:

25 XII1

- Groups of formula XIII, as disclosed for example in WO 2006/052457
A2 and WO 2006/118345A1:

wherein

20 R¹¹-R¹⁸

are independently of each other selected from the group consisting of H, halogen, alkyl, substituted alkyl, aryl, substituted aryl, heteroaryl, substituted heteroaryl, -CN, -CHO, -COR²⁰, - CR²⁰=NR¹⁹, - OR²⁰, - SR²⁰, -SO₂R²⁰, -POR²⁰R¹⁹, -PO₃R²⁰, -OCOR²⁰, -CO₂R²⁰, - NR²⁰R¹⁹, -N=CR²⁰R¹⁹, -NR²⁰COR¹⁹, and -CONR²⁰R¹⁹; and any of R¹¹⁻¹⁸ do optionally form a ring system with adjacent repeating units in the polymer; and R¹⁷ and R¹⁸ do optionally together form a ring system;

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R¹⁹ and R²⁰ are independently of each other selected from the group consisting of H, alkyl, substituted alkyl, aryl, substituted aryl, heteroaryl, and substituted heteroaryl; and adjacent R^{19,20} groups do optionally together form a ring system;

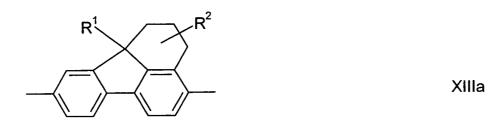
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and wherein either
a) R¹⁷ forms a ring system with R¹⁶ or

b) R¹⁷ forms a ring system with R¹⁶, and R¹⁸ forms a ring system with R¹¹, wherein the two ring systems do optionally share more than one atom.

The groups of formula XIII are preferably of the following subformula:

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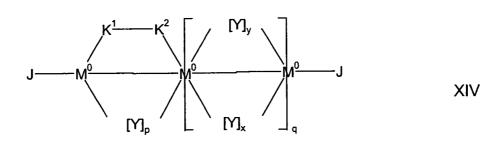


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wherein R¹ and R² are as defined above.

- Groups of formula XIV, as disclosed for example in DE 102006003710.3:

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wherein

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M⁰ is in case of multiple occurrence independently of one another an aromatic, heteroaromatic or non-aromatic ring system having 2 to 40 C atoms, which is unsubstituted or substituted with one or more identical or different groups R¹,

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 $K^{1,2}$ and Y are in case of multiple occurrence independently of one another a bridging group forming a cyclic sytem with M, selected from $B(R^1)$, $C(R^1)_2$, $Si(R^1)_2$, C=O, C=S, C=Se, C=Te, $C=NR^1$, $C=C(R^1)_2$, O, S, S=O, SO_2 , $S(R^1)_2$, $N(R^1)$, $P(R^1)$, $P(=O)R^1$, $P(=S)R^1$, C=C or a combination or one, two, three or four of these groups,

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- J is a linkage group to the polymer, and may also be a substituted oder unsubstituted C-C-double or triple bond, a substituted aromatic or heteroaromatic or non-aromatic cyclic ring system having 2 to 40 C atoms, which is unsubstituted or substituted with one or more identical or different groups R¹,
- R¹ is as defined in formula I,
- 10 x, y, p are in case of multiple occurrence independently of one another 0 or 1, with the proviso that at least one of x and y is 1,
 - q is an integer ≥ 1 .

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The groups of formula XIV are preferably selected from the following subformulae:

wherein R1 is defined as above.

Further preferred emissive groups A^{em} are those selected from the following formulae:

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 Vinyltriarylamines of formula XV, as disclosed for example in DE 10 2005 060473.0:

wherein

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Ar¹¹ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²¹,

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Ar¹² is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²²,

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Ar¹³ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²³,

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Ar¹⁴ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²⁴,

30

y¹¹ is in case of multiple occurrence independently of one another selected from H, F, Cl or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and optionally two groups Y¹¹, or a group Y¹¹ and an adjacent group R²¹, R²⁴, Ar¹¹ or Ar¹⁴, do together form an aromatic, mono- or polycyclic ring system,

35

denote in case of multiple occurrence independently of one another H, halogen, -CN, -NC, -NCO, -NCS, -OCN, -SCN, -C(=O)NR⁰R⁰⁰, -C(=O)X⁰, -C(=O)R⁰, -NH₂, -NR⁰R⁰⁰, -SH, -SR⁰,

-SO₃H, -SO₂R⁰, -OH, -NO₂, -CF₃, -SF₅, optionally substituted silyl, or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and wherein optionally two or more groups R²¹-²⁴ do together form an aliphatic or aromatic, mono- or polycyclic ring system; and wherein R²¹, R²² and R²³ may also denote a covalent bond in a polymer,

5

 X^0 , R^0 and R^{00} are as defined in formula I.

10

- is in each occurrence independently of one another 1, 2 or 3, i
- is in each occurrence independently of one another 1, 2 or 3, k

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is in each occurrence independently of one another 0 or 1. 0

The groups of formula XV are preferably selected from the following subformulae:

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wherein V is a covalent bond in a polymer, v is 0 or 1, and R⁵ has one of the meanings of R²⁴ given above. The benzene rings are optionally substituted by one or more groups R⁵.

The ratio of units of formula XV in polymer 2 is preferably from 0.1 to 10 mol %, very preferably from 0.5 to 5 mol % and most preferably from 1 to 2.5 mol %.

1,4-Bis(2-thienylvinyl)benzenes of formula XVI, as disclosed for example in WO 2005/030827:

$$\begin{array}{c|c}
S & R^1 \\
Ar & S \\
R^2 & XVI
\end{array}$$

wherein R¹ and R² are as defined in formula I and Ar has one of the

meanings of Ar¹ given in formula II.

The groups of formula XVI are preferably of the following subformula:

5 (L")_r

wherein L" and r are as defined above, and L" is preferably alkyl, aryl, perfluoroalkyl, thioalkyl, cyano, alkoxy, heteroaryl, alkylaryl, or arylalkyl, very preferably H, phenyl, C₁₋₁₂-alkyl or -alkoxy.

Very preferred groups of formula XVI are of the following subformula:

wherein L" is preferably alkyl, aryl, perfluoroalkyl, thioalkyl, cyano, alkoxy, heteroaryl, alkylaryl, or arylalkyl, very preferably H, phenyl, C₁₋₁₂-alkyl or -alkoxy.

 1,4-Bis(2-arylenvinyl)benzenes of formula XVII, as disclosed for example in WO 00/46321:

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wherein r and R are as defined above and u is 0 or 1.

The groups of formula XVII are preferably selected from the following subformulae:

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wherein L" is preferably alkyl, aryl, perfluoroalkyl, thioalkyl, cyano, alkoxy, heteroaryl, alkylaryl, or arylalkyl, very preferably H, phenyl, C₁₋₁₂-alkyl or -alkoxy; and L" is H, F, Cl, CN or optionally fluorinated linear, branched or cyclic alkyl having 1 to 20 C atoms, or optionally substituted aryl having 5 to 30 C atoms, preferably H or phenyl.

Groups of formula XVIII:

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$$X^{21}$$
 XVIIII

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wherein

is O, S, SO₂ C(R^x)₂ or N-R^x wherein R^x is aryl or substituted aryl or aralkyl with 6 to 40 C atoms, or alkyl with 1 to 24 C atoms, preferably aryl with of 6 to 24 C atoms, very preferably alkylated aryl with 6 to 24 C atoms,

Ar²¹ is optionally substituted aryl or heteroaryl with 6 to 40, preferably 6 to 24, very preferably 6 to 14 C atoms.

10 - Groups of the following formulae:

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$$R^{25}$$
 R^{26} N N N N $-(Ar^{22})_{a1}$ X^{22} $(Ar^{23})_{b1}$ XIX

25 wherein

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is R²⁷C=CR²⁷ or S, wherein preferably each R²⁷ is independently selected from hydrogen, alkyl, aryl, perfluoroalkyl, thioalkyl, cyano, alkoxy, heteroaryl, alkylaryl, or arylalkyl,

R²⁵ and R²⁶ are the same or different and are each a substituent group,

Ar²² and Ar²³ are in case of multiple occurrence independently of one another a bivalent aromatic or heteroaromatic ring

system having from 2 to 40 C atoms, which is optionally substituted by one or more groups R^{25} , and

a1 and b1 are in each occurrence independently of one another 0 or 1.

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- Groups of the following formulae:

$$X^{23}$$
 $XXIB$

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wherein X^{23} is NH, O, or S.

- Groups of the following formulae:

20 R R XXII

25

R R XXIII

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- 57 -

wherein R and R' are as defined above, and are in each occurrence independently of one another preferably H, alkyl, aryl, perfluoroalkyl, thioalkyl, cyano, alkoxy, heteroaryl, alkylaryl, or arylalkyl. R is preferably H, phenyl, or alkyl having 1, 2, 3, 4, 5 or 6 C atoms. R' is preferably n-octyl or n-octyloxy;

- Groups selected from the following formulae:

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wherein Ph denotes phenyl;

Further suitable and preferred groups that can be used as exciton formation groups A^{ex} or emissive groups A^{em} are those selected from the following formulae:

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- Triazines of formula XXXIII (as disclosed for example in WO 01/49769):

$$Ar^{24} - N(Ar^{25})_2 \qquad XXXIII$$

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wherein Ar²⁴ is a substituted or unsubstituted heteroaryl group and each Ar²⁵ is the same or different and comprises a substituted or unsubstituted aryl or heteroaryl group. Preferred groups of this type are the following:

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10 wherein R' and R" are as defined above;

 Triarylamines of formula XXXIV comprising one or more heteraromatic groups (as disclosed for example in WO 03/35714):

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$$-Ar^{26}-Ar^{27}-N(Ar^{28})-Ar^{29}-Ar^{30}-$$
 XXXIV

wherein Ar²⁶⁻³⁰ have independently of each other one of the meanings of Ar¹ given above, and at least one of Ar²⁶⁻³⁰ is optionally substituted heteroaryl, for example thiophene or triazine. Preferred groups of this type are the following:

wherein R' and R" are as defined above and are preferably alkyl, per-fluoroalkyl, alkylaryl, arylalkyl, heteroaryl, aryl, alkoxy, aryloxy or thioalkyl.

Further suitable and preferred backbone groups are those selected from the following formulae, as disclosed for example in WO2006/114364A1 and EP1345477A2:

$$R^{36}$$

$$R^{36}$$

$$R^{36}$$

$$R^{37}$$

$$R^{38}$$

$$R^{39}$$

$$R^{34}$$

$$R^{34}$$

Wherein R³¹⁻³⁶ are independently of each other selected from H, alkyl, aryloxy, an aromatic rest, a condensed aromatic ring system, a heteroaromatic rest, -CH=CH(E)- or (Z)-CH=CH-C₆H₅, acryloyl, methacryloyl, methylstyryl, -O-CH=CH2, or glycidyl,

In a further preferred embodiment of the present invention polymer 2 comprises one or more identical or different emissive groups A^{em} emitting light from the triplett state. Suitable groups of this type are known to the

expert and are described in the literature. Especially preferred are groups based on or derived from metal complexes.

These groups are preferably selected of the formula $M(L)_z$, wherein M is a metal atom, L is in each occurrence independently of one another an organic ligand that is bonded to or coordinated with M via one, two or more positions, and z is an integer > 1, preferably 1, 2, 3, 4, 5 or 6, and wherein these groups are linked to the polymer via one or more, preferably one, two or three positions, preferably via the ligands L.

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M is a metal atom preferably selected from transition metals, especially those of group VIII, or lanthanoides, very preferably selected from Rh, Os, Ir, Pt, Au, Sm, Eu, Gd, Tb, Dy, Re, most preferably Os, Ir, Rh or Pt. M may also denote Al, Be or Zn.

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L is preferably a mono- or bidentate organic ligand which can be neutral (non-ionic) or anionic. Such ligands are known in the art. Suitable neutral monodentate ligands are for example CO, isonitriles, amines, phosphines, phosphites, arsines, stilbines, or N-heterocycles like pyridine, pyridazine, pyrazine or triazine. Suitable anionic monodentate ligands are for example halogenides, cyanide, cyanates, isocyanates, thiocyanates, isothiocyanates, alcoholates, thioalcoholates, amides, carboxylates, or anionic N-heterocycles like pyrrolide, imidazolide, pyrazolide. Suitable bidentate ligands are for example diamines like ethylenediamine or its N-alkylated derivatives, imines, diimines, heterocycles with two N atoms like 2,2'-bipyridine or ophenathroline, diphosphines, 1,3-diketonates derived from 1,3-diketones like acetylacetone, 3-ketonates derived from 3-keto esters, carboxylates derived from amino carboxylic acids like pyridine-2-carboxylic acid, quinoline-2carboxylic acid, picolinic acid (pyridine-2-carboxylic) acid, salicyl iminates derived from salicyl imines, dialcoholates derived from dialcoholes like ethylene glycol or dithiolates derived from dithioles like 1,2-ethylene dithiolate. Further suitable ligands are selected from mono- or polycyclic aromatic or heteroaromatic groups having 4 to 50 C atoms, preferably containing at least one N atom, which are optionally substituted, like for example 8-quinolinol, benzoquinolinol, 2-phenyl pyridine, 2-phenyl benzothiazole, 2-phenyl-benzoxazole, porphyrine or their derivatives.

Suitable and preferred groups of this type are for example those of the following formulae (as disclosed for example in WO 02/68435):

 $\begin{array}{c|c}
\hline
 & (R^1)_n & XXXV \\
\hline
 & (R^1)_n & (XX)
\end{array}$

15 $(R^{1})_{n}$ (YY) H R^{3} R^{3} (XX) R^{3}

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XXXVIII

M3 5

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$$M = \begin{pmatrix} (R^1)_n \\ (XX) \end{pmatrix}$$

XXXIX

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 (R^1) , M.

XXXX

wherein R¹, R³, m and n are as defined above, M is a metal atom as defined above, preferably Rh or Ir, XX is a linkage in the polymer and YY is in each occurrence independently of one another O, S or Se.

Further suitable and preferred groups of this type are for example those as disclosed in US 6,696,180 or US 2002/0193532 A1:

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wherein R has one of the meanings of R¹ given above, and one or more, preferably one, two or three groups R, very preferably one group at one, two or three of the ligands, denotes a linkage to neighbouring units in the polymer.

Polymer 1 is preferably selected of the following formulae

$$\begin{array}{c|c} \hline A^h \\ \hline n2 \\ \hline \end{array} \begin{array}{c} B \\ \hline \end{array} \begin{array}{c} \\ \hline n1 \\ \hline \end{array}$$
 P11

$$-\begin{bmatrix} B-A^h B \end{bmatrix}_{m1}$$
 P13

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$$- \left[-A^{h} B - A^{h} \right]_{m_{1}}$$
 P14

wherein

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A^h is a hole transporting group as defined above, B is a backbone group as defined above, PCT/EP2007/005722

m1 is an integer > 1,

0 < n1 < 1,

0 < n2 < 1,

n2 < n1, and

5 preferably n1+n2 = 1.

Preferably A^h in formulae P11-14 is selected of formulae II and III, and B is selected of formulae I, IV, V, VI, VII, IX, X, XII, XIII and XIV.

Further preferably polymer 1 is a copolymer comprising one or more sections selected of formulae P11, P12, P13 and P14.

Polymer 2 is preferably selected of the following formulae

$$-[B-A^e-B]_{n2}$$
 $-[B-A^{ex}-B]_{n3}$ $-[B-A^{em}-B]_{n4}$ $-[B-A^{em}-B]_{n4}$

wherein

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A^e is an electron transporting group as defined above,

A^{ex} is an exciton formation group as defined above,

A^{em} is an emissive group as defined above,

B is a backbone group as defined above,

0 < n1 < 1.

0 < n2 < 1

0 < n3 < 1,

 $0 \le n4 < 1$

preferably n1 > each of n2, n3 and n4, and preferably n1+n2+n3+n4 = 1.

A^{ex} may additionally have emissive property. A^{em} may additionally have exciton formation property. Preferably the groups A^e, A^{ex}, A^{em} and B are selected from formulae I-XVII as indicated above.

In addition to the groups disclosed above, the polymers of the present invention may comprise one or more units which are typically used as backbone units or as blue emitters in light-emitting polymers. These are generally units which comprise at least one aromatic or otherwise conjugated group, but do not shift the emission wavelength into green or red. Preferred are aromatic groups having 4 to 40 C atoms (but also stilbene or tolane derivatives, and some bis(styrene)arylene derivatives) including, but not limited to, substituted or unsubstituted 1,4-phenylene, 1,4-naphthylene, 1,4- or 9,10-anthracenylene, 2,7- or 3,6phenanthrenylene, 4,4'-biphenylene, 4,4"-terphenylene, 4,4'-bi-1,1'naphthylene, 4,4'-stilbene, 4,5-dihydropyrene derivatives, 4,5,9,10tetrahydropyrenes derivatives (e.g. as disclosed in EP 0 699 699), fluorene derivatives (e.g. as disclosed in EP 0 842 208, WO 99/54385, WO 00/22027, WO 00/22026, WO 00/46321), spirobifluorene derivatives (e.g. as disclosed in EP 0 707 020, EP 0 894 107, WO 03/020790, WO 02/077060) and 5,7-dihydrodibenzoxepine derivatives, furthermore so-

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In a further preferred embodiment of the present invention polymer 2 comprises one or more identical or different exciton formation units (dyes), which upon absorbing photons, for example from sun light, form excitons, which further dissociate by transferring holes to polymer 1 and electrons to the electron transporting unit in polymer 2. Preferred groups of this type are, perylene derivates as disclosed for example in Angew. Chem. Int. Ed. 2006, 45, 3364-3368, and Ruthenium dyes and teir derivatives, as disclosed for example in Nature 1991, 353, pp737 and Angew.Chem. Int. Ed. 2005, 44, 5740-5744.

called "Ladder-PPPs" (LPPP) (e.g. as disclosed in WO 92/18552), and PPPS containing ansa structures (e.g. as disclosed in EP 0 690 086).

- The polymers of the present invention may be statistical or random copolymers, alternating or regionegular copolymers, block copolymers or combinations thereof. They may comprise two, three or more distinct monomer units.
- The polymers of the present invention may be prepared by any suitable method. For example, they can be suitably prepared by aryl-aryl coupling

reactions, such as Yamamoto coupling, Suzuki coupling, Stille coupling, Sonogashira coupling or Heck coupling. Suzuki coupling and Yamamoto coupling are especially preferred.

The monomers which are polymerised to form the repeat units of the polymers of the present invention can be prepared according to suitable methods which are known to the expert and have been disclosed in the literature. Suitable and preferred methods for the preparation of the indenofluorene monomers of formula I are described for example in WO 2004/041901. Suitable and preferred methods for the preparation of the triarylamine monomers of formula II are described for example in WO 99/54385. Suitable and preferred methods for the preparation of the phenanthrene monomers of formula IV are described for example in WO 2005/104264 A1. Suitable and preferred methods for the preparation of the vinyltriarylamine monomers of formula XV are described for example in DE102005060473.0.

Preferably the polymers are prepared from monomers comprising one of the above mentioned groups, which are linked to two polymerisable groups P. Accordingly, for example the monomers for the units of formula I1 are selected of the following formula

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wherein P is a polymerisable group and R¹ is as defined above. The monomers for the units of the other formulae disclosed above are built accordingly.

Preferably the groups P are independently of each other selected from CI, Br, I, O-tosylate, O-triflate, O-mesylate, O-nonaflate, SiMe_{3-z}F_z (wherein z is 1 or 2), O-SO₂Z, B(OZ¹)₂, -CZ²=C(Z²)₂, -C \equiv CH and Sn(Z³)₃, wherein Z and Z¹⁻³ are selected from the group consisting of alkyl and aryl, each being

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optionally substituted, and two groups Z¹ may also form a cyclic group.

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Preferred methods for polymerisation are Suzuki polymerisation, as described for example in WO 00/53656, Yamamoto polymerisation, as described in for example in T. Yamamoto et al., Progress in Polymer Science 1993, 17, 1153-1205 or in WO 2004/022626 A1, and Stille coupling. For example, when synthesizing a linear polymer by Yamamoto polymerisation, a monomer as described above having two reactive halide groups P is preferably used. When synthesizing a linear polymer by Suzuki polymerisation, preferably a monomer as described above is used wherein at least one reactive group P is a boron derivative group.

Suzuki polymerisation may be used to prepare regioregular, block and random copolymers. In particular, random copolymers may be prepared from the above monomers wherein one reactive group P is halogen and the other reactive group P is a boron derivative group. Alternatively, block copolymers or alternating copolymers, in particular AB-type copolymers, may be prepared from a first and a second of the above monomers wherein both reactive groups of the first monomer are boron and both reactive groups of the second monomer are halide. The synthesis of block copolymers is described in detail for example in WO 2005/014688 A2.

It is also possible to prepare e.g. an AB-type polymer from a single monomer unit having the structure P-AB-P.

Suzuki polymerisation employs a Pd(0) complex or a Pd(II) salt. Preferred Pd(0) complexes are those bearing at least one phosphine ligand such as Pd(Ph₃P)₄. Another preferred phosphine ligand is tris(ortho-tolyl)phosphine, i.e. Pd(o-Tol)₄. Preferred Pd(II) salts include palladium acetate, i.e. Pd(OAc)₂. Suzuki polymerisation is performed in the presence of a base, for example sodium carbonate, potassium phosphate or an organic base such as tetraethylammonium carbonate. Yamamoto polymerisation employs a Ni(0) complex, for example bis(1,5-cyclooctadienyl) nickel(0).

35 As alternatives to halogens as described above, leaving groups of formula -O-SO₂Z can be used wherein Z is as described above. Particular

examples of such leaving groups are tosylate, mesylate and triflate.

A further aspect of the present invention is an electronic or electrooptical device comprising one or more polymer blends as described above and below. A further aspect is the use of the polymer blends as described above and below in an electronic or electrooptical device. Especially preferred devices are PLEDs.

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The device assembly typically consists of a substrate (like e.g. a glass sheet 10 or a plastic foil), a first electrode, an interlayer comprising a conductive, doped polymer, a semiconductor layer according to the invention, and a second electrode. The device is patterned and provided with contacts according to the desired application and then sealed, in order to avoid contact with water and air which could drastically reduce its lifetime. It may 15 also be preferred to use a conductive, electrically doped polymer as electrode, in which case the interlayer comprising the conductive polymer can be omitted. For use in OFETs and TFTs the device has to contain a further electrode (gate electrode) in addition to the first and second electrode (source and drain electrode). The gate electrode is separated from the 20 organic semiconductor layer by an insulator layer comprising a dielectric material having a dielectric constant that is usually high (but in some cases may also be low). It may also be suitable that the device comprises one or more further layers depending on the desired application.

The electrodes are selected such their potential matches the potential of the adjacent organic layer, to ensure that hole or electrode injection is as efficient as possible. Preferred cathode materials are metals with low electronic work function, metal alloys or multilayered structures comprising different metals, such as alkaline earth metals, alkaline metals, main group metals or lanthanoides (e.g. Ca, Ba, Mg, Al, In, Mg, Yb, etc.). In case of multilayered structures it is also possible to use further metals in addition to the above-mentioned metals, which have a relatively high electronic work function, like e.g. Ag. In such cases normally combinations of metals are used, like e.g. Ca/Ag or Ba/Ag. It may also be preferred to apply a thin intermediate layer of a material with a high dielectric constant between the metallic cathode and the organic semiconductor. Useful materials for this

purpose are e.g. alkaline or alkaline earth metal fluorides or oxides (e.g. LiF, Li₂O, BaF₂, MgO, NaF etc.). The thickness of this dielectric layer is preferably from 1 to 10 nm.

Preferred anode materials are those having a high electronic work function. Preferably the anode has a potential of more than 4.5 eV (vs. vaccum). Suitable materials for this purpose are metals having a high redox potential, like e.g. Ag, Pt or Au. It may also be preferred to use metal/metaloxide electrodes (e.g. Al/Ni/NiO_x, Al/Pt/PtO_x). For some applications at least one of the electrodes has to be transparent, in order to enable e.g. irradiation of the organic material (in OSCs) or decoupling of light (in OLEDs/PLEDs, O-LASERs). A preferred assembly comprises a transparent anode. Preferred anode materials for this purpose are conductive mixed metal oxides. Especially preferred are Indium Tin Oxide (ITO) or Indium Zinc Oxide (IZO). Further preferred are also conductive, doped organic materials, especially conductive doped polymers.

As charge injection layer on the anode a variety of doped conductive polymers can be used. Preferred polymers are those having a conductivity of > 10⁻⁸ S/cm. The potential of the layer is preferably 4 to 6 eV (vs. vacuum). The layer thickness is preferably from 10 to 500 nm, very preferably from 20 to 250 nm. Very preferably derivatives of polythiophene, like poly(3,4-ethylenedioxy-2,5-thiophene) (PEDOT) and polyaniline (PANI) are used. Doping is usually carried out with acids or oxidative agents. Preferably doping is carried out with polymeric or polymer bound Brönsted acids. Preferred materials for this purpose are polymeric sulfonic acids, especially polystyrene sulfonic acid, polyvinyl sulfonic acid and poly-(2-acrylamido-2-methyl-propane sulfonic acid) (PAMPSA). The conductive polymer is usually applied as an ageous solution or dispersion and is insoluble in organic solvents, which allows to apply the subsequent layer from organic solution.

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Typically, the blend is coated on a device structure which consists of ITO-coated glass which was covered by a layer of conductive polymer. The thickness of the conductive polymer can vary between 10 and 200 nm depending on ITO-roughness. The polymer blend is then coated from

solution onto the conductive polymer in a thickness varying from 20 to 120 nm, preferably from 60 to 100 nm. Typically, the conductive polymer and the blend is baked after coating to remove rest of solvents (water and organic solvent) for a certain period of time. Baking temperatures vary from the polymers used and are typically in the range of 100-200 °C for 1-120 minutes, preferably 130-200 °C for 1-60 minutes and most preferably 150-180°C for 10-30 minutes.

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After coating the cathode on top of the blend, the device is typically covered by a lid to avoid penetration of moisture and oxygene.

In another embodiment of this invention the blend can also be coated onto an interlayer. In this case a polymer having hole transport property, which may be polymer 1 or a different polymer, is coated from organic solution onto the conductive polymer and the film is heated at elevated temperature to form an interlayer. Then, non-solidified residues of the interlayer are washed away with the organic solvent and the blend is coated directly onto the deposited interlayer. Alternatively, the blend can also be directly coating onto the interlayer. After coating the blend, a heating procedure as described above is applied again.

The electronic or electrooptical device can also be for example an organic field effect transistor (OFET), thin film transistor (TFT), organic solar cell (O-SC), organic laser diode (O-laser), organic integrated circuit (O-IC), radio frequency identification (RFID) tag, photodetector, sensor, logic circuit, memory element, capacitor, charge injection layer, Schottky diode, planarising layer, antistatic film, conducting substrate or pattern, photoconductor, electrophotographic element, or organic light emitting transistor (OLET).

Another aspect of the invention relates to a solution comprising one or more polymer blends as described above and below and one or more organic solvents.

Examples of suitable and preferred organic solvents include, without limitation, dichloromethane, trichloromethane, monochlorobenzene, o-

dichlorobenzene, tetrahydrofuran, anisole, morpholine, toluene, o-xylene, m-xylene, p-xylene, 1,4-dioxane, acetone, methylethylketone, 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, ethyl acetate, n-butyl acetate, dimethylformamide, dimethylacetamide, dimethylsulfoxide, tetraline, decaline, indane, methyl benzoate, ethyl benzoate, mesitylene and/or mixtures thereof.

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The concentration of the polymers in the solution is preferably 0.1 to 10% by weight, more preferably 0.5 to 5% by weight. Optionally, the solution also comprises one or more binders to adjust the rheological properties, as described in WO 2005/055248 A1.

After the appropriate mixing and ageing, solutions are evaluated as one of the following categories: complete solution, borderline solution or insoluble. The contour line is drawn to outline the solubility parameter-hydrogen bonding limits dividing solubility and insolubility. 'Complete' solvents falling within the solubility area can be chosen from literature values such as published in "Crowley, J.D., Teague, G.S. Jr and Lowe, J.W. Jr., Journal of Paint Technology, 38, No 496, 296 (1966)". Solvent blends may also be used and can be identified as described in "Solvents, W.H.Ellis, Federation of Societies for Coatings Technology, p9-10, 1986". Such a procedure may lead to a blend of 'non' solvents that will dissolve both the polymers of the present invention, although it is desirable to have at least one true solvent in a blend.

- It is desirable to generate small structures or patterns in modern microelectronics to reduce cost (more devices/unit area), and power consumption. Patterning of the layer of the invention may be carried out for example by photolithography or electron beam lithography.
- For use as thin layers in electronic or electrooptical devices the polymer blends or solutions of the present invention may be deposited by any suitable method. Liquid coating of devices such OLEDs is more desirable than vacuum deposition techniques. Solution deposition methods are especially preferred. Preferred deposition techniques include, without limitation, dip coating, spin coating, ink jet printing, letter-press printing, screen printing, doctor blade coating, roller printing, reverse-roller printing,

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offset lithography printing, flexographic printing, web printing, spray coating, brush coating or pad printing. Ink-jet printing is particularly preferred as it allows high resolution displays to be prepared.

- Selected solutions of the present invention may be applied to prefabricated device substrates by ink jet printing or microdispensing. Preferably industrial piezoelectric print heads such as but not limited to those supplied by Aprion, Hitachi-Koki, InkJet Technology, On Target Technology, Picojet, Spectra, Trident, Xaar may be used to apply the organic semiconductor layer to a substrate. Additionally semi-industrial heads such as those manufactured by Brother, Epson, Konica, Seiko Instruments Toshiba TEC or single nozzle microdispensers such as those produced by Microdrop and Microfab may be used.
- In order to be applied by ink jet printing or microdispensing, the polymers should be first dissolved in a suitable solvent. Solvents must fulfil the requirements stated above and must not have any detrimental effect on the chosen print head. Additionally, solvents should have boiling points >100°C, preferably >140°C and more preferably >150°C in order to prevent
 operability problems caused by the solution drying out inside the print head. Apart from the solvents methoned above, suitable solvents include substituted and non-substituted xylene derivatives, di-C₁₋₂-alkyl formamide, substituted and non-substituted anisoles and other phenol-ether derivatives, substituted heterocycles such as substituted pyridines, pyrazines, pyrimidines, pyrrolidinones, substituted and non-substituted *N,N*-di-C₁₋₂-alkylanilines and other fluorinated or chlorinated aromatics.

A preferred solvent for depositing a polymer according to the present invention by ink jet printing comprises a benzene derivative which has a benzene ring substituted by one or more substituents wherein the total number of carbon atoms among the one or more substituents is at least three. For example, the benzene derivative may be substituted with a propyl group or three methyl groups, in either case there being at least three carbon atoms in total. Such a solvent enables an ink jet fluid to be formed comprising the solvent with the polymer, which reduces or prevents clogging of the jets and separation of the components during spraying. The

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solvent(s) may include those selected from the following list of examples: dodecylbenzene, 1-methyl-4-tert-butylbenzene, terpineol limonene, isodurene, terpinolene, cymene, diethylbenzene. The solvent may be a solvent mixture, that is a combination of two or more solvents, each solvent preferably having a boiling point >100°C, more preferably >140°C. Such solvent(s) also enhance film formation in the layer deposited and reduce defects in the layer.

The ink jet fluid (that is mixture of solvent, binder and semiconducting compound) preferably has a viscosity at 20°C of 1-100mPa's, more preferably 1-50mPa's and most preferably 1-30mPa's.

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The polymers or solutions according to the present invention can additionally comprise one or more further components like for example surface-active compounds, lubricating agents, wetting agents, dispersing agents, hydrophobing agents, adhesive agents, flow improvers, defoaming agents, deaerators, diluents which may be reactive or non-reactive, auxiliaries, colourants, dyes or pigments, sensitizers, stabilizers, or inhibitors.

Unless the context clearly indicates otherwise, as used herein plural forms of the terms herein are to be construed as including the singular form and vice versa.

Throughout the description and claims of this specification, the words "comprise" and "contain" and variations of the words, for example "comprising" and "comprises", mean "including but not limited to", and are not intended to (and do not) exclude other components.

It will be appreciated that variations to the foregoing embodiments of the invention can be made while still falling within the scope of the invention. Each feature disclosed in this specification, unless stated otherwise, may be replaced by alternative features serving the same, equivalent or similar purpose. Thus, unless stated otherwise, each feature disclosed is one example only of a generic series of equivalent or similar features.

All of the features disclosed in this specification may be combined in any

combination, except combinations where at least some of such features and/or steps are mutually exclusive. In particular, the preferred features of the invention are applicable to all aspects of the invention and may be used in any combination. Likewise, features described in non-essential combinations may be used separately (not in combination).

It will be appreciated that many of the features described above, particularly of the preferred embodiments, are inventive in their own right and not just as part of an embodiment of the present invention. Independent protection may be sought for these features in addition to or alternative to any invention presently claimed.

The invention will now be described in more detail by reference to the following examples, which are illustrative only and do not limit the scope of the invention.

Unless stated otherwise, all specific values of physical parameters like efficiency, VIL curves and lifetime, as given above and below refer to a temperature of 25° C (+/- 1° C). Ratios of monomers or repeating units in polymers are given in mol %. Ratios of polymers in polymer blends are given in weight %. The molecular weight of polymers is given as weight average molecular weight M_{w} (GPC, polystyrene standard).

Example 1: Polymers

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The following polymers are synthesized by Suzuki coupling as disclosed in WO03/048225.

Polymer1 is the copolymer of following monomers:

Polymer2a is the copolymer of following monomers:

Polymer2b is the copolymer of following monomers:

Polymer2c is the copolymer of following monomers:

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Polymer2d is the copolymer of following monomers:

Example 2: cyclovoltammetry measurement on the polymers

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The HOMO levels of the above polymers are measured by cyclovoltammetry (CV). The CV measurements are conducted in dichlormethane solution using tetrabutylammonium chloride as conducting salt, Ag/AgCl/KCl (3mol/l) as reference electrode, Au as working electrode and Pt as counter electrode. Figure 1, figure 2, figure 3 and figure 4 are the CV curves for poylmer1, polymer2a, polymer2c and polymer2d. 3 cycles were measured for every polymer. The HOMO levels were calculated by the average voltage of the local maximum peak in oxidation-curve and corresponding local niminum peak reduction-curve. Care should be taken to find the correct local minima and maxima, for example change the axis case to find all peaks. The results are summarised in Table 1, with the alignments of the different function groups to the corresponding peaks.

For polymer1, polymer2a and polymer2c, all peaks are very clearly spoted. All there cycles give essentially the same results. For polymer2d, the peak 2 is not very clearly to seen. This is due to the very strong axidation and reduction for the neighbour peak 3. However, according to the shoulder in

oxidation curve (see also the area marked with the dashed ellipse), an estimation could still be made for peak 2. Therefore, very reliable HOMO levels are obtained by CV measurement.

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	CV HOMO [eV]	Corresponding Group in polymer	Function of the group
Polymer1	36分数60%(智慧	进步的 经通过通过 医克里克氏征	
Peak 1	-5,14	M2-M1-M2	Hole transporting unit
Polymer2a	1967年的 在 1967年	为19.65 (1.45) [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45] [1.45]	的一个加工的现在分词是对于有" 没会 "的
Peak 1	-4,84	M4-M5-M4	Exciton formation unit
Peak 2	-5,15	M4-M3-M4	Electron transporting unit
Polymer2c		数"等有"的"全全 的"的现在是 "学习"的,在一种,但是	以下,1925年后的 对政治会 的
Peak 1	-4,87	M6-M5-M6	Exciton formation unit
Peak 2	-5,18	M6-M7-M6	Emissive units
Peak 3	-5,58	M6-M3-M6	Electron transport unit
Polymer2d	生物。其實物的政治	对于特别的证据的 "是是我们的是是一个人的。"	
Peak 1	-4,85	M6-M5-M6	Exciton formation unit
Peak 2	-5,29	M6-M8-M6	Emissive unit
Peak 3	-5,59	M6-M3-M6	Electron transport unit

Example 3: Quantum simulation on energy levels of the polymers

As mentioned before, the applicants established a very reliable quantum simulation method to determine the energy levels of organic materials. The HOMO and LUMO levels of individual units are calculated by Gaussian 03W using the DFT method. In the calculation, the trimer M2-M1-M2 is taken as hole transport unit in polymer1; M4-M3-M4 as electron transport unit and M4-M5-M4 as both exciton formation and emissive unit in polymer2a; M6-M3-M6 as electron transport unit and M6-M5-M6 as both exciton formation and emissive unit in polymer2b; M6-M3-M6 as electron transport unit, M6-M5-M6 as exciton formation unit and M6-M7-M6 as emissive unit in polymer2c; M6-M3-M6 as electron transport unit, M6-M5-M6 as exciton formation unit and M6-M8-M6 as emissive unit in polymer2d.

The CV measured and DFT calculated energy levels for the different function units in the polymers are shown in Table 2. It shold be pointed out that the LUMO values calculated by DFT are less precise than the HOMO values. Therefore the LUMO values listed in Table 1 should be taken as indicative rather than quantitative, but still confirm the claimed properties.

Table 2:

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Calculated trimer	Corresponding unit	HOMO Calibrated	LUMO Calibrated	HOMO measured
by DFT	in polymer	[eV]	[eV]	by CV [eV]
	Hole transport unit	100	2000年度	47.000
M2-M1-M2	in polymer1	-5.14	-2.47	-5.14
	Electron transport units			\$1980 D
M4-M3-M4	in polymer2a	-5.19	-2.55	5.15
	Electron transport units	577	-27-12-12-12-12-12-12-12-12-12-12-12-12-12-	1.4%
M6-M3-M6	in polymer2b, 2c,2d	-5.55	-2.58	-5.58 ¹⁾
	Exciton formation unit			
M4-M5-M4	in polymer2a	-4.89	-2.29	-4.84
	Exciton formation unit		Mark the	Saldkardary :
M6-M5-M6	in polymer2b, 2c, 2d	-4.93	- 🌣 -2.29	-4:86 ¹⁾
	Emissive units			Granda in
M6-M7-M6	in polymer2c	-5.17	2.57	5.18
	Emissive units		国本名式(单名	
M6-M8-M6	in polymer2d	-5.27	-2.88	5.29

¹⁾ Average of polymer2c and polymer2d

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The CV measurement on the electron transport units of polymer2a and polymer2b-d is performed on polymers with 50%M4/50%M3 and 50%M6/50%M3. The HOMO of exciton formation unit in polymer2b-d is measured on a polymer of 50%M6/40%M3/10%M5. CV measurements on emissive units have not been performed. As can be seen in Table 2, the calculation on HOMO level agrees with the measurement very well. Therefore, the established simulation method can be used as a reliable tool for designing the material system according to the present invention. Later on, polymer1 will be used as interlayer polymer and hole transporting component in blends, and polymer2a-2d will be used as emissive and electron transporting component in blends. It should be pointed out that the HOMO of the electron transport unit of polymer2a is just 0.01eV by CV and 0.05eV by DFT lower than the HOMO of the hole transport unit of polymer1. In contrast, the HOMO of the electron transport unit of polymer2b-2d is about 0.41eV by DFT and 0.44eV by CV lower than the HOMO of the hole transport unit of polymer1. This difference is essential for the different performance of the blend systems.

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Thus, a blend of polymer 1 with either of polymers 2b, 2c or 2d represents a blend according to the present invention. In contrast, a blend of polymer 1 with polymer 2a is not a blend according to the present invention and will be

used for comparison purposes as described below.

Example 4: Device preparation

- 5 PLED devices with an interlayer are prepared according to the following procedure:
 - 1) Depositing 80nm PEDOT (Baytron P AI 4083) onto indium tin oxide coated glass substrate by spin coating.
 - 2) Depositing 20nm polymer1 by spin coating from toluene solution having a concentration of 5% wt/l.
 - 3) Heating the device at 180°C for 1 hour.
 - 4) Depositing the emissive layer by spin-coating from toluene solution to a thickness of around from 60 to 80nm.
 - 5) Depositing by evaporation over the emissive layer a cathode Ba/Al
- 15 6) Encapsulation of the device.

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For a device without interlayer the steps 2 and 3 are omitted.

Example 5 (Comparison Example): Light Emitting Diodes using blends of polymer1 and polymer2a as emissive layer

Blends of 5%, 10% and 15 weight% polymer1, respectively, with polymer2a are tested as emissive layer (80nm thickness) in a PLED device without interlayer. The reference is a device using polymer1 as interlayer and pure polymer2a as emissive layer (80nm thickness). Table 3 shows the comparison of the performance of these devices.

Table 3:

Con. Polymer1 in blends	Max. Eff. [cd/A]	Uon [V]	U @ 100Cd/m ² [V]	CIE @ 100 cd/m²	Lifet [hrs @	
Reference	3.44	2.97	4.11	0.16 / 0.22	443	400
2% Polymer1	2.36	3.38	5.26	0.18 / 0.31	66.4	400
5%Polymer1	2.00	4.1	5.1	0.17 / 0.27	81	400
10%Polymer1	2.49	3.7	4.6	0.16 / 0.24	159	400
15%Polymer1	2.59	4.1	5.1	0.17 / 0.25	170	400

Figure 5 shows the lifetime curves of the PLEDs using blends of polymer1 and polymer2a, in comparison with the reference. It can be seen that no

blend PLED can give a comparable performance as the reference, and that the decay behaviors of the blend PLEDs are very different than that of the reference.

- This shows that if the HOMO gap between the hole transporting unit of the first polymer and the electron transporting unit of the second polymer is too small, no working blend system with satisfactory performance can be obtained.
- 10 <u>Example 6: Light Emitting Diodes using blends of polymer1 and polymer2b</u> as emissive layer

Blend of 15 weight% polymer1 with polymer2b was tested as emissive layer in a PLED device without an interlayer. The reference is a device using polymer1 as interlayer and pure polymer2b 65nm thick as emissive layer.

Table 4 shows the summary of the performance of PLEDs using blend of 15 wt% polymer1 and polymer2b, where EQE stands for external quantum efficiency.

Table 4:

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	Max. Eff.	Uon	U(100)	CIE @	EQE@	LT (OC
	[cd/A]	[V]	[V]	100 cd/m ²	Max. Eff.	[hrs @	nits]
Reference	4,78	3,4	5,7	0,15 / 0,16	3,76%	107	1000
15% polymer1	3,72	3,3	5,9	0,15 / 0,16	2,91%	90	1000

In Figure 6, the lifetime curves are compared. The PLED using blend as emissive layer without the additional interlayer shows a comparable lifetime to the reference, though the efficiency is a little bit lower.

30 Example 7: Light Emitting Diodes using blends of polymer1 and polymer2c as emissive layer

Blends of 2%, 10%, 5% and 20 weight% polymer1, respectively, with polymer2c are tested as emissive layer in a PLED device without an interlayer. The reference is a device using polymer1 as interlayer and pure polymer2a 65nm thick as emissive layer.

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Table 5 shows the summary of the performance of PLEDs using blends of polymer1 and polymer2c in different concentration, where EQE stands for external quantum efficiency.

Table 5:

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Con. polymer1 in	Thickness of	Max. Eff.	Uon	U(100)	CIE @	EQE @	LT C	C
blends	emissive layer	[cd/A]	[V]	[V]	100 cd/m ²	Max. Eff.	[hrs @	nits]
Reference	65nm	5.92	3.4	5.7	0.15 / 0.17	4.41%	304	1001
0% polymer1	65nm	4.15	3.7	6.5	0.15 / 0.18	2.96%	14	999
2%polymer1	65nm	5.10	3.5	6.1	0.15 / 0.17	3.75%	25	1000
10% polymer1	65nm	4.95	3.1	5.2	0.15 / 0.16	3.83%	133	1000
15%polymer1	65nm	4.14	3.2	5.2	0.15 / 0.15	3.36%	201	1000
15%polymer1	80nm	4.67	3.4	5.8	0.15 / 0.17	3.38%	346	1000
20%polymer1	65nm	3.70	3.0	4.7	0.15 / 0.15	3.02%	122	1000

In Figure 7, the lifetime curves are compared. It can be seen that all blends have much better lifetime than the pure polymer2c as emissive layer in a single layer device, which has only 14 hrs lifetime. Compared to the reference, all single layer blends PLEDs give very similar CIE coordinates. The EQE and efficiency is slightly lower than reference. Surprisingly, a PLED with a blend comprising 15 weight% of polymer1 has a lifetime that is even superior to the reference interlayer device.

Example 8: Light Emitting Diodes using blends of polymer1 and polymer2c as emissive layer in interlayer device

A blend of polymer1 and polymer2c (15:85) is tested as emissive layer in a PLED device comprising an interlayer of polymer1, in comparison to a PLED interlayer device using pure polymer2c as emissive layer. The results are summarized in Table 6.

30 <u>Table 6:</u>

Emissive layer	Max. Eff. [cd/A]	Uon [V]	U(100) [V]	CIE @ 100 cd/m²	LT [hrs @	
65nm Polymer2c on IL	5.92	3.4	5.7	0.15 / 0.17	304	1001
80nm Blend on IL	3.96	3.5	6.0	0.15 / 0.20	470	1000

Figure 8 clearly shows that using a polymer blend according to the invention as emissive layer in an interlayer device can still significantly improve the lifetime, in this case from 304 hrs to 470hrs.

5 Example 9: Light Emitting Diodes using blends of polymer1 and polymer2d as emissive layer in single layer device and interlayer device

A blend of 15%wt polymer1 with 85%wt polymer2d is tested as emissive layer in a PLED device with and without an interlayer (of polymer1). The reference is a PLED device using polymer1 as interlayer and pure polymer2d in a thickness of 65nm as emissive layer. The results are summarized in Table 7.

Table 7:

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Emissive Layer	Thickness	Max. Eff.	Uon	U(100)	CIE@	EQE @	LT	DC
1	of LEP	[cd/A]	[V]	[V]	100 cd/m²	Max. Eff.	[hr:	s @
IL/Polymer2d	65nm	17.41	3.2	4.9	0.31 / 0.59	5.31%	404	5993
Blend polymer1 and 2d	80nm	12.96	3.0	4.8	0.31 / 0.59	3.96%	365	5898
IL/Blend polymer1 and 2d	80nm	12.29	3.1	5.1	0.32 / 0.59	3.78%	681*	5999

* 487hrs @ 57%, predicted as 681hrs

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In Figure 9, the lifetime curves are compared. The polymer blend shows a slightly worse performance, i.e. efficiency and lifetime, than the reference. However, if the blend is tested in an interlayer device, it gives a much longer lifetime (681hrs) than the reference (404hrs).

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Example 10: Light Emitting Diodes using polymer2b and polymer2c as emissive layer in interlayer device

Pure polymer2b and polymer2c are tested as emissive layer in a PLED device with an interlayer (of polymer1). The thickness of polymer2b and polymer2c are 65nm. The results are summarized in Table 8.

Table 8:

	Max. Eff.	Üon [V]	Ú(100) [Ý]	CIE @ :/ 100 cd/m²	EQE @ Max. Eff.	LT [hrs @	DC ၨ୬ nits]
Polymer2b	4,78	3,4	5,7	0,15 / 0,16	3,76%	107	1000
Polymer2c	5,92	3,4	5,7	0,15 / 0,17	4,41%	304	1001

In Figure 10, the lifetime curves are compared. In summary, the polymer with an emissive unit (polymer2c) shows a much better performance, especially lifetime, than the polymer only with exciton formation unit, e.g. without additional emissive unit.

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Patent Claims

1. Polymer blend comprising

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- a first polymer comprising a unit having hole transporting property (hole transporting unit),
- a second polymer comprising a unit having electron transporting property (electron transporting unit) and a unit having exciton formation property (exciton formation unit), and optionally comprising one or more further units having emissive property (emissive unit) and/or exciton formation property,
- wherein each of said units has a HOMO ("highest occupied molecular orbital") and a LUMO ("lowest unoccupied molecular orbital"), and the difference between said HOMO and LUMO is hereinafter referred to as the "energy gap",
- wherein the HOMO of both said hole transporting unit of said first polymer and said exciton formation unit of said second polymer are at least 0.2 eV higher than the HOMO of said electron transporting unit of said second polymer,
 - with the proviso that a blend that comprises a polymer comprising substituted or unsubstituted fluorene and triarylamine units and further comprises a polymer comprising substituted or unsubstituted fluorene and benzothiadiazol units is excluded.
- 2. Polymer blend according to claim 1, characterized in that the HOMO of both said hole transporting unit of the first polymer and said exciton formation unit of the second polymer are at least 0.3 eV higher than the HOMO of said electron transporting unit of the second polymer.
- 3. Polymer blend according to claim 1 or 2, characterized in that the HOMO of said exciton formation unit of the second polymer is at least 0.4 eV higher than the HOMO of said electron transporting unit of the second polymer.
- 4. Polymer blend according to one or more of claims 1 to 3,
 35 characterized in that the LUMO of said exciton formation unit of the second polymer is higher than the LUMO of said electron transporting

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unit of the second polymer.

- 5. Polymer blend according to one or more of claims 1 to 4, characterized in that said second polymer comprises at least one unit having emissive property.
- 6. Polymer blend according to one or more of claims 1 to 5, characterized in that the exciton formation unit of said second polymer is itself an emissive unit.

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- 7. Polymer blend according to one or more of claims 1 to 6, characterized in that said second polymer contains one or more additional emissive units, wherein the excitation energy is transferred from exciton formation unit to each of said additional emissive units, and the energy of each of said additional emissive units is smaller than the energy gap of said exciton formation unit.
- 8. Polymer blend according to claim 8, characterized in that said additional emissive units emit blue, green and/or red light.

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- 9. Polymer blend according to one or more of claims 1 to 8, characterized in that the ratio of said exciton formation unit is from 0.01 to 10 mol%.
- 25 10. Polymer blend according to one or more of claims 1 to 9, characterized in that the ratio of said additional emissive units is from 0.01 to 10 mol%.
- 11. Polymer blend according to one or more of claims 1 to 10, 30 characterized in that the second polymer has a negligible hole mobility compared to the first polymer.
 - 12. Polymer blend according to one or more of claims 1 to 11, characterized in that the first polymer has a negligible electron mobility compared to the second polymer.

- 13. Polymer blend according to one or more of claims 1 to 12, characterized in that the proportion of polymer 1 in the blend is from 0.1 to 50 weight%.
- 5 14. Polymer blend according to one or more of claims 1 to 13, characterized in that the first polymer comprises one or more further emissive units.
 - 15. Polymer blend according to claim 14, characterized in that said further emissive units have a lower HOMO than the HOMO of said hole transporting unit of the first polymer and a higher LUMO than the LUMO of said electron transporting unit of the second polymer, and accept excitation energy from said exciton formation unit of the second polymer by energy transfer.
- 15 16. Polymer blend according to one or more of claims 1 to 15, characterized in that the first and/or second polymer comprises one or more identical or different groups of formula I

$$(Ar^{1})_{a}$$

$$(Ar^{2})_{b}$$

$$(Br^{1})_{a}$$

25 wherein

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- A, B and B' are independently of each other, and in case of multiple occurrence independently of one another, a divalent group, preferably selected from -CR¹R²-, -NR¹-, -PR¹-, -O-, -S-, -SO-, -SO₂-, -CO-, -CS-, -CSe-, -P(=O)R¹-, -P(=S)R¹- and -SiR¹R²-,
- are independently of each other identical or different groups selected from H, halogen, -CN, -NC, -NCO, -NCS, -OCN, -SCN, -C(=O)NR⁰R⁰⁰, -C(=O)X, -C(=O)R⁰, -NH₂, -NR⁰R⁰⁰, -SH, -SR⁰, -SO₃H, -SO₂R⁰, -OH, -NO₂, -CF₃, -

SF₅, optionally substituted silyl, or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and optionally the groups R¹ and R² form a spiro group with the fluorene moiety to which they are attached,

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X is halogen,

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R⁰ and R⁰⁰ are independently of each other H or an optionally substituted carbyl or hydrocarbyl group optionally comprising one or more hetero atoms,

each g is independently 0 or 1 and each corresponding h in the same subunit is the other of 0 and 1,

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m is an integer ≥ 1 ,

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Ar¹ and Ar² are independently of each other mono- or polynuclear aryl or heteroraryl that is optionally substituted and optionally fused to the 7,8-positions or 8,9-positions of the indenofluorene group,

a and b are independently of each other 0 or 1,

- 25 17. Polymer blend according to one or more of claims 1 to 16, characterized in that the ratio of groups of formula I in the first polymer is from 10 to 80 mol%.
- 18. Polymer blend according to one or more of claims 1 to 17,

 characterized in that the first polymer comprises one or more identical or different groups of formula II

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wherein

Y is N, P, P=O, PF₂, P=S, As, As=O, As=S, Sb, Sb=O or Sb=S,

5 Ar¹ which may be the same or different, denote, independently if in different repeat units, a single bond or an optionally substituted mononuclear or polynuclear aryl group,

Ar² which may be the same or different, denote, independently if in different repeat units, an optionally substituted mononuclear or polynuclear aryl group,

Ar³ which may be the same or different, denote, independently if in different repeat units, an optionally substituted mononuclear or polynuclear aryl group, which may be optionally substituted by a bridging group linking different chain residues of formula II, and

m is 1, 2 or 3,

- 20 19. Polymer blend according to one or more of claims 1 to 18, characterized in that the ratio of groups of formula II in the first polymer is from 10 to 80 mol%.
- 20. Polymer blend according to one or more of claims 1 to 19,
 25 characterized in that the second polymer comprises one or more identical or different groups of formula IV

$$\begin{array}{c|c}
R^{1} & R^{2} \\
\hline
 & Ar^{6} X^{1} \downarrow g \downarrow h
\end{array}$$
IV

wherein

R¹ and R² have independently of each other one of the meanings

given in claim 16,

 X^1 and X^2 are independently of each other -CR¹=CR¹-, -C=C- or -N-Ar⁸-,

- Ar⁶⁻⁸ are in case of multiple occurrence independently of one another a bivalent aromatic or heteroaromatic ring system having from 2 to 40 C atoms, which is optionally substituted by one or more groups R¹ as defined in claim 16,
- 10 g is in each occurrence independently of one another 0 or 1,
 - h is in each occurrence independently of one another 0, 1 or 2.
- 21. Polymer blend according to one or more of claims 1 to 20,
 15 characterized in that the second polymer comprises one or more identical or different groups of formula VI

$$(R^{1})_{n}$$

$$(R^{1})_{n}$$

$$(R^{2})_{m}$$

$$(R^{2})_{m}$$

$$(R^{2})_{m}$$

wherein

- X is in each occurrence independently of one another CH, CR¹ or N,
- 30 Z is in each occurrence independently of one another a single bond, CR^5R^6 , CR^5R^6 - CR^5R^6 , $CR^5=CR^6$, O, S, N-R⁵, C=O, C=CR⁵R⁶ or SiR⁵R⁶;
- R^{1,2} have independently of each other the meanings given in claim 11, 35
 - R^{5,6} have independently of each other one of the meanings of R¹,

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- m is in each occurrence independently of one another 0, 1, 2, or 3,
- n is in each occurrence independently of one another 0, 1, 2, 3 or 4.

22. Polymer blend according to one or more of claims 1 to 21, characterized in that the second polymer comprises one or more identical or different groups of formula XV

wherein

Ar¹¹ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²¹,

20 Ar¹² is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²²,

Ar¹³ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²³,

Ar¹⁴ is in each occurrence independently of one another mono-or polycyclic aryl or heteroaryl, which is optionally substituted by one or more groups R²⁴,

y¹¹ is in case of multiple occurrence independently of one another selected from H, F, Cl or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and optionally two groups Y¹¹, or a group Y¹¹ and an adjacent group R²¹, R²⁴, Ar¹¹ or Ar¹⁴, do

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together form an aromatic, mono- or polycyclic ring system,

denote in case of multiple occurrence independently of one another H, halogen, -CN, -NC, -NCO, -NCS, -OCN, -SCN, -C(=O)NR⁰R⁰⁰, -C(=O)X⁰, -C(=O)R⁰, -NH₂, -NR⁰R⁰⁰, -SH, -SR⁰, -SO₃H, -SO₂R⁰, -OH, -NO₂, -CF₃, -SF₅, optionally substituted silyl, or carbyl or hydrocarbyl with 1 to 40 C atoms that is optionally substituted and optionally comprises one or more hetero atoms, and wherein optionally two or more groups R²¹⁻²⁴ do together form an aliphatic or aromatic, mono- or polycyclic ring system; and wherein R²¹, R²² and R²³ may also denote a covalent bond in a polymer,

X⁰, R⁰ and R⁰⁰ are as defined in claim 16,

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- i is in each occurrence independently of one another 1, 2 or 3,
- k is in each occurrence independently of one another 1, 2 or 3,

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- o is in each occurrence independently of one another 0 or 1.
- 23. Polymer blend according to one or more of claims 1 to 22, characterized in that the second polymer comprises one or more identical or different groups of formula XVI

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wherein R¹ and R² are as defined in claim 11 and Ar has one of the meanings of Ar¹ given in claim 12.

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24. Polymer comprising an electron transporting unit and an exciton formation unit, and optionally comprising one or more further emissive

units and/or exciton formation units, wherein each of said units has a HOMO and a LUMO, and the HOMO of said first exciton formation unit is at least 0.3 eV higher than the HOMO of said electron transporting unit.

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- 25. Polymer according to claim 24, characterized in that it is a second polymer as defined in one or more of claims 3 to 10 and 20 to 23.
- 26. Use of a polymer blend or a polymer according to one or more ofclaims 1 to 26 in an electronic or electrooptical device.
 - 27. Electronic or electrooptical device comprising a polymer blend or a polymer according to one or more of claims 1 to 26.
- 15 28. Electronic or electrooptical device according to claim 27, wherein said device comprises a conductive polymer layer and/or a hole transporting layer, and the polymer blend is coated directly onto said conductive polymer and/or hole transport layer.
- 29. Electronic or electrooptical device according to claim 27, wherein said device does not comprise a conductive polymer or hole transporting layer.
- 30. Electronic or electrooptical device according to one or more of claims 27 to 29, which is a polymer light emitting diode (PLED), organic field effect transistor (OFET), thin film transistor (TFT), organic solar cell (O-SC), organic laser diode (O-laser), organic integrated circuit (O-IC), radio frequency identification (RFID) tag, photodetector, sensor, logic circuit, memory element, capacitor, charge injection layer, Schottky diode, planarising layer, antistatic film, conducting substrate or pattern, photoconductor, electrophotographic element, or organic light emitting transistor (OLET).
 - 31. Method for preparing a polymer blend, by mixing a first polymer with a second polymer as defined in one or more of claims 1 to 23, and optionally with one or more further polymers.

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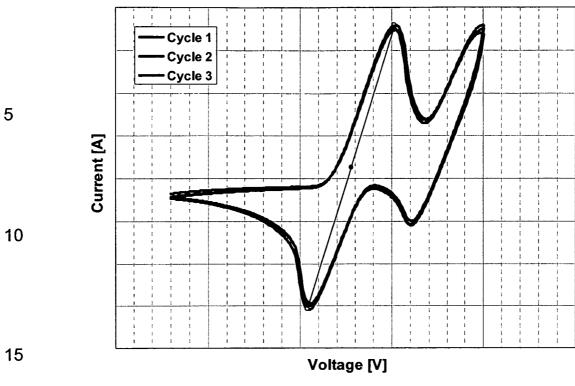
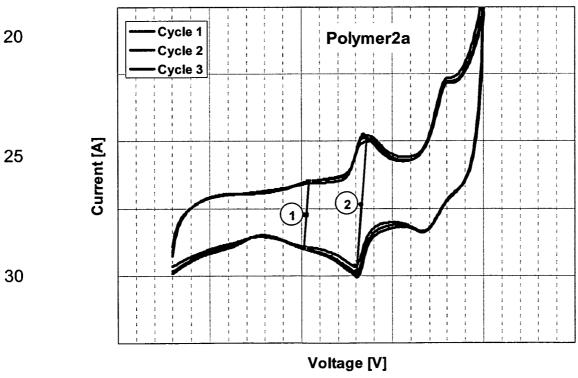
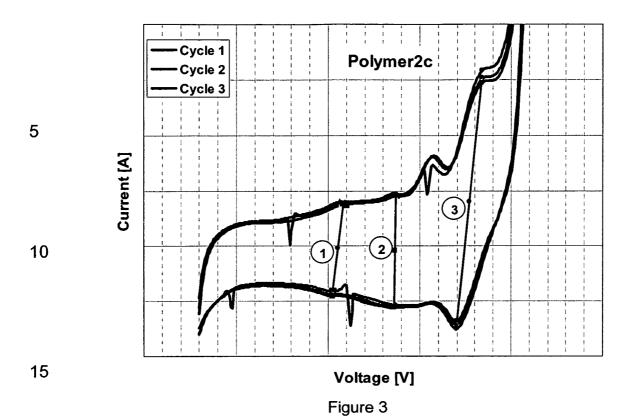


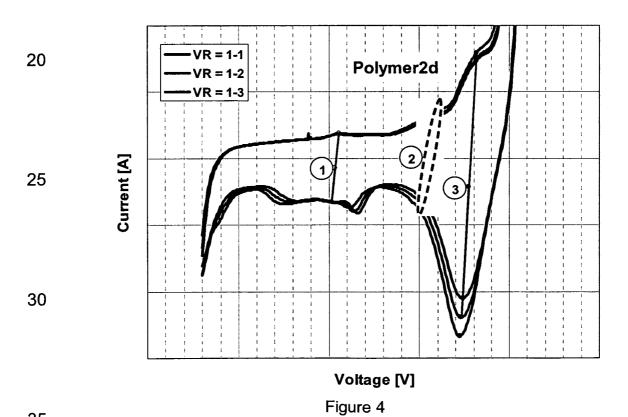
Figure 1



35 Figure 2

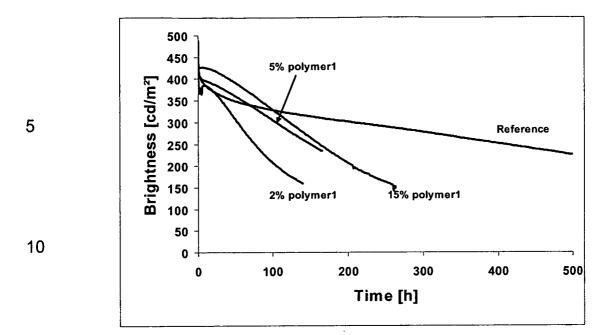
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- 3/5 -



15 Figure 5

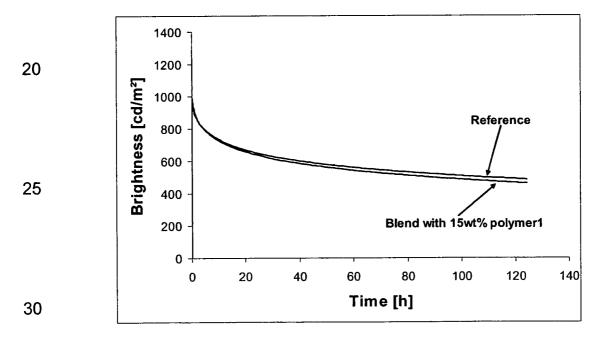


Figure 6

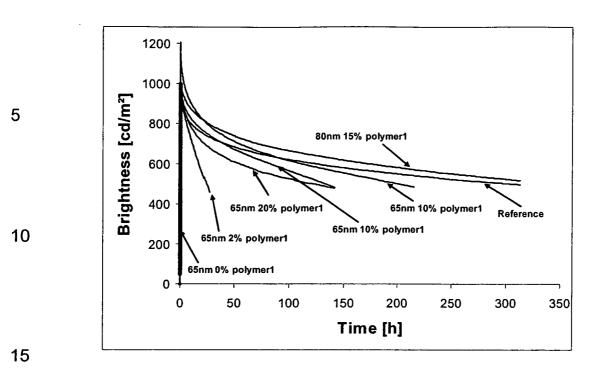


Figure 7

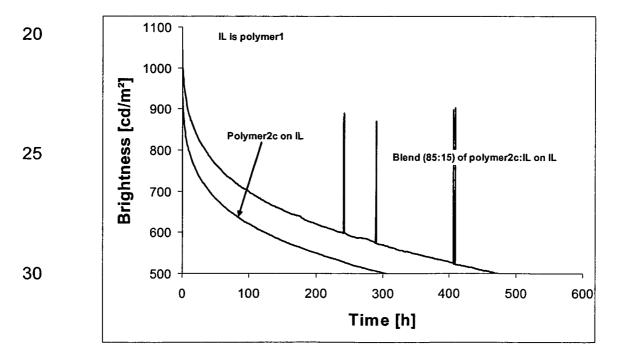
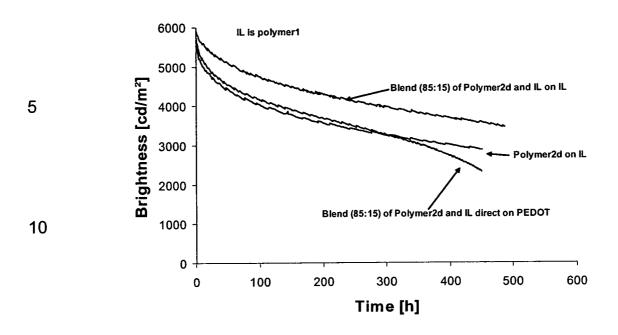
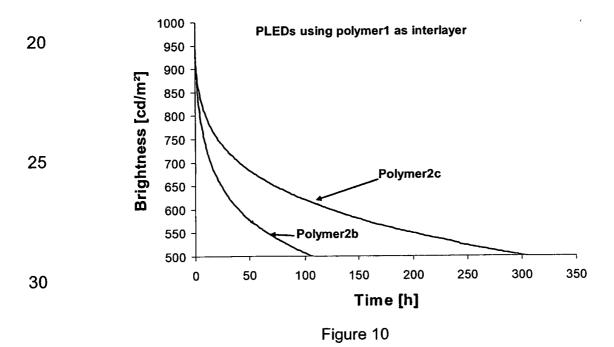


Figure 8



15 Figure 9



INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/005722

A. CLASSIFICATION OF SUBJECT MATTER
INV. H01L51/00 H01L5 C08L65/00 H01L51/50 H01B1/12 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 C08L H01B Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data C. DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Category* Υ WO 2005/053052 A (CAMBRIDGE DISPLAY TECH 1-23. [GB]; PATEL NALINKUMAR [GB]; HALLS 26 - 31JONATHAN [GB) 9 June 2005 (2005-06-09) cited in the application page 15 - page 20; claims 1-21; figures 2,3 US 2001/026878 A1 (WOO EDMUND P [US] ET Υ 1-23.AL) 4 October 2001 (2001-10-04) 26 - 31paragraph [0019] - paragraph [0046]; example 1; table 1 Y WO 99/48160 A (CAMBRIDGE DISPLAY TECH 1-23, [GB]; BURROUGHES JEREMY HENLEY [GB]; 26-31 FRIEND RIC) 23 September 1999 (1999-09-23) cited in the application claims 1-55; table 4 -/--X Further documents are listed in the continuation of Box C. See patent family annex. Special categories of cited documents : "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the "A" document defining the general state of the art which is not considered to be of particular relevance earlier document but published on or after the international *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another "Y" document of particular relevance; the claimed invention citation or other special reason (as specified) cannot be considered to involve an inventive step when the document is combined with one or more other such docu-"O" document referring to an oral disclosure, use, exhibition or ments, such combination being obvious to a person skilled document published prior to the international filing date but later than the priority date claimed "&" document member of the same patent family Date of the actual completion of the international search Date of mailing of the international search report 12 November 2007 19/11/2007 Authorized officer Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Marsitzky, Dirk Fax: (+31-70) 340-3016

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/005722

C(Continua	tion). DOCUMENTS CONSIDERED TO BE RELEVANT	
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Υ	MORTEANI, DHOOT, KIM, SILVA, GREENHAM, MURPHY, MOONS, CINA, BURROUGHES, FRIEND: "Barrier-Free Electron-Hole Capture in Polymer Blend Heterojunction Light-Emitting Diodes" ADVANCED MATERIALS, vol. 15, no. 20, 16 October 2003 (2003-10-16), - 2003 pages 1708-1712, XP002452116 cited in the application page 1711; figure 1	1-23, 26-31
X	WO 00/55927 A (CAMBRIDGE DISPLAY TECH [GB]; TOWNS CARL ROBERT [GB]; DELL RICHARD O [G) 21 September 2000 (2000-09-21) page 20 - page 25; example 6	24,25
X	US 2003/045642 A1 (WU WEISHI [US] ET AL) 6 March 2003 (2003-03-06) examples 1-3	24,25
		
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International application No. PCT/EP2007/005722

INTERNATIONAL SEARCH REPORT

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)
This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:
Claims Nos.: because they relate to subject matter not required to be searched by this Authority, namely:
Claims Nos.: because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
Claims Nos.: because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).
Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)
This International Searching Authority found multiple inventions in this international application, as follows:
see additional sheet
1. As all required additional search fees were timely paid by the applicant, this international search report covers allsearchable claims.
2. As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.
3. As only some of the required additional search fees were timely paid by the applicant, this international search reportcovers only those claims for which fees were paid, specifically claims Nos.:
4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:
The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee. The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-23, 26 (part)-30 (part), 31

Polymer blend comprising a) a first polymer comprising a hole transport unit and b) a second copolymer having b1) an exciton formastion unit and b2) an electron transporting unit wherein c) both the HOMO of the hole transport unit and the exciton fomation unit are at least 0.2 eV higher than the HOMO of the electron transport unit. Use of the polymer blend in optoelectronic devices. Method for preparing the polymer blend via mixing.

2. claims: 24-25, 26 (part) - 30 (part)

Polymer comprising a) an electron transporting unit and b) an exciton formation unit wherein c) the HOMO of the exciton formation unit is at least 0.3 eV higher than the HOMO of the electron transport unit. Use of this polymer in optoelectronic devices.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2007/005722

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
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US 2003045642	A 1	06-03-2003	NON	E	



专利名称(译)	聚合物共混物及其在有机发光器件中的用途		
公开(公告)号	EP2044636A1	公开(公告)日	2009-04-08
申请号	EP2007764910	申请日	2007-06-28
申请(专利权)人(译)	MERCK PATENT GMBH		
当前申请(专利权)人(译)	MERCK PATENT GMBH		
[标]发明人	PAN JUNYOU MEYER FRANK		
发明人	PAN, JUNYOU MEYER, FRANK		
IPC分类号	H01L51/00 H01L51/50 C08L65/00	H01B1/12	
CPC分类号	C08L65/00 C08G61/02 C08G61/10 C08G61/12 C08G61/126 C08G2261/314 C08G2261/3142 C08G2261/3162 C08G2261/5222 C08L2205/02 C09B57/008 C09B57/10 H01L51/0036 H01L51/0039 H01L51/0043 H01L51/5012 Y02E10/549 Y10T428/31786		
优先权	2006015459 2006-07-25 EP		
外部链接	Espacenet		

摘要(译)

本发明涉及包含一种或多种空穴传输聚合物和一种或多种电子传输聚合物的新型聚合物共混物,这些共混物在电子和电光器件中,特别是在有机发光二极管(OLED)中的用途,以及包含该共混物的OLED。聚合物混合物。