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# (54) Luminescent element material and luminescent element comprising the same

Luminiszentes Elementmaterial und lumineszentes Element damit

Matériau d'élément luminescent et élément luminescent le comportant

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## Description

### Technical Field

**[0001]** The present invention relates to light emitting device materials and to a light emitting device which can convert electrical energy into light and can be used for display elements, flat panel displays, back lights, lighting, interiors, signs, signboards, electronic cameras, light signal generators and the like.

### Technical Background

**[0002]** There has recently been considerable research into organic multi-layered thin film light-emitting devices which emit light when electrons injected from a cathode and holes injected from an anode recombine within an organic fluorescent body interposed between the electrodes. Such devices are a focus of attention on account of their characteristics of thin shape, high luminance at. low driving voltage and polychromic light emission based on suitable selection of the fluorescent materials.

**[0003]** Numerous research organizations have been carrying out such research since C.W. Tang and co-workers at Kodak first described the fact that an organic multi-layered thin film element emits light of high luminance (Appl. Phys. Lett. 51(12) 21, p.913, 1987). A typical organic multi-layered thin film light-emitting element construction proposed by the Kodak research group is that in which there are provided, in turn, on an ITO glass substrate, a hole transporting diamine compound, 8-hydroxyquinoline aluminium as the emissive layer and Mg:Ag as the cathode, and 1,000 cd/m$^2$ green coloured light emission is possible at a driving voltage of about 10 V.

**[0004]** In this organic multi-layered·thin film light-emitting device structure, as well as the aforesaid anode/hole transporting layer/emissive layer/cathode, there may also be suitably provided an electron transporting layer. The hole transporting layer has the function of transporting the holes injected from the anode to the emissive layer, while the electron transporting layer transports the electrons injected from the cathode to the emissive layer. By interposing these layers along with the emissive layer between the two electrodes, the luminance efficiency and the durability are enhanced. As examples of device structures employing these, there are structures comprising an anode/hole transporting layer/emissive layer/electron transporting layer/cathode and an anode/emissive layer/electron transporting layer/cathode, etc.

**[0005]** However, many conventional emissive materials, hole transporting materials and electron transporting materials lack durability, and crystallization takes place due to the heat evolved from the device by the prolonged passage of current, so that the device life is shortened.

**[0006]** In particular, taking the case of the electron transporting material, with many of the existing materials there are problems such as the desired emission colour not be obtained for reasons such as interaction with the emissive material or there being admixed light emission by the electron transporting material itself, while even where highly efficient emission is obtained the durability is poor. In US Patent 5393614, a specific phenanthroline derivative is used as the electron transporting material, but while highly efficient emission is shown there is crystallization during prolonged operation, and the thin film turns cloudy. Quinolinol metal complexes and benzoquinolinol metal complexes are also materials which show comparatively good characteristics in terms of luminance efficiency and durability but, since these materials themselves have a high emissive capacity in the blue-green to yellow region, when employed as electron transporting materials there is admixed emission from these materials themselves and the chromatic purity is adversely affected.

**[0007]** The present invention has the objective of resolving such problems of the prior-art and offering a light emitting device which is excellent in its thermal stability, has high luminance efficiency, high luminance and excellent chromatic purity.

### Disclosure of the Invention

**[0008]** The light emitting device of the present invention relates to a light emitting device which is characterized by claim 1.

**[0009]** The electron transporting layer of the present invention is the layer into which electrons are injected from the cathode and which transports the electrons, and it is desirable that the electron injection efficiency be high and that the injected electrons are highly efficiently transported. However, taking into account the hole and electron transportation balance, in the case where its role is primarily to efficiently block the flow of holes from the anode to the cathode without recombination, then, even if the electron transporting capacity of the electron transporting layer is not all that high, its effect in terms of enhancing the luminance efficiency will be the same as that of a material with a high electron transportation capacity. Consequently, electron transporting layer in the present invention will encompass within the same definition a hole blocking layer which can efficiently inhibit the transport of holes.

**[0010]** The material from which the electron transporting layer of the present invention is composed is an organic compound of molecular weight at least 400. With an organic compound of molecular weight less than 400, the electron transporting layer will be thermally unstable and readily crystallize, so that stable emission is not obtained in terms of prolonged operation. A molecular weight of at least 600 is further preferred.

**[0011]** The material from which the electron transporting layer of the present invention is composed has a glass transition temperature of at least 90°C, more preferably at least 120°C and still more preferably at least 150°C. Furthermore, a film of a compound of high cold crystallization temperature does not readily crystallize, and it is preferred that the cold crystallization temperature be at least 140°C, more preferably at least 170°C and still more preferably at least 200°C. Moreover, it is desirable that it be a compound where no cold crystallization temperature is observed. Reference here to no cold crystallization temperature being observed means that when the glass transition temperature and the cold crystallization temperature of a .sample are measured, no clear cold crystallization temperature is found when the temperature of the sample is raised at a certain fixed rate. The glass transition temperature and the cold crystallization temperature are measured using a differential scanning calorimeter based on temperature-modulated DSC.

**[0012]** The ionization potential of the material from which the electron transporting layer of the present invention is composed, will be at least 5.9 eV. When the ionization potential is at least 5.9 eV, it is possible to prevent very efficiently the holes injected from the anode from flowing to the cathode side without recombining within the emissive layer, so the luminance efficiency is enhanced. Furthermore, since the electron transporting layer itself does not fluoresce, light emission of high chromatic purity is obtained only from the emissive layer. More preferably, the value is at least 6.0 eV. The absolute value of the ionization potential will differ according to the measurement method but, in the present invention, measurement is carried out with an atmospheric air type UV photoelectron analyzer (AC-1, produced by the Riken Keiki Co. Ltd) using a thin film evaporated onto an ITO glass substrate.

**[0013]** Moreover it is also preferred that the ionization potential of the electron transporting layer of the present invention be at least 0.1 eV greater than the ionization potential of the emissive layer. In the case where there is a difference in ionization potential of at least 0.1 eV between the adjacent electron transporting layer and emissive layer, it is possible to efficiently prevent the holes injected from the anode from flowing to the cathode side without recombining within the emissive layer. In terms of a high temperature operational environment, it is further preferred that this difference be at least 0.15 eV and still more preferably at least 0.2 eV. The difference in the ionization potentials in the present invention is calculated from the ionization potentials of the respective individual layers measured on their own by the aforesaid method. Moreover, the value of the ionization potential will change with the state of the sample. Consequently, in the case where the emissive layer or the electron transporting layer is a mixed layer comprising two or more materials, the value of the ionization potential of this mixed layer is measured.

**[0014]** The organic compound from which the electron transporting layer is composed preferably contains a plurality of parent skeletal structures having an electron transporting capacity, with this plurality of parent skeletal structures being connected together by connecting units. The benzoquinoline structure is the parent skeletal structure with an electron transporting capacity. In addition to having an electron transporting capacity, a high ionization potential is obtained with the benzoquinoline structure. These benzoquinoline structures are provided with a functional group with an electron transporting capacity such as the vinyl group, carbonyl group, carboxyl group, aldehyde group, nitro group, cyano group, halogen, sulphone, phosphorus oxide or the like. Of these, phosphorus oxide is preferred.

**[0015]** The organic compounds from which the emissive layer and the electron transporting layer are composed are preferably compounds which can sublime. Here, 'can sublime' means that when heated in a vacuum they volatilize without decomposition, and so thin-film formation is possible. The light emitting device of the present invention has a multi-layered structure so, where there is employed an organic compound which can sublime, it is possible to form a multi-layered structure readily using a dry process such as vacuum vapour deposition. Again, in the case of the formation of a doping layer within the emissive layer, by employing the method of co-evaporation along with the host material or the method of simultaneous evaporation after prior-mixing with the host material, it is possible to form the doping layer with an outstanding degree of control. Furthermore, in a display device where display is effected by the matrix or segment system, it is necessary to obtain emission in a desired pattern and organic compounds which can sublime can readily be patterned by a dry process.

**[0016]** The electron transporting layer of the present invention is not necessarily restricted to one type of aforesaid organic compound and a plurality of such materials may be mixed together or provided as a multilayer. In the case of a multilayer, the aforesaid parameters may be satisfied by the single layer adjacent to the emissive layer. Furthermore, with the objective of enhancing the transportation capacity of the electron transporting layer as a whole, or with the objective of enhancing the thermal or electrochemical stability, the electron transporting layer may be formed by adding, to the electron transporting material, an organic compound, inorganic compound or metal complex which does not have an electron transporting capacity.

**[0017]** The emissive layer is the layer where the emissive substance is actually formed, and the emissive substance may be composed of one type of organic compound or there may be employed a mixed layer comprising two or more types of organic compound. From the point of view of enhancing the luminance efficiency, chromatic purity and durability, the emissive layer is preferably composed of two or more types of organic compound. An example of a combination of two or more types of organic compound is the combination of a host material and a dopant material. In such circumstances, the host material primarily has the emissive layer thin-film forming ability and carrier transporting capacity, while the dopant material primarily has the emission ability. As emission mechanisms, there are the energy transfer type and the carrier trap type. In the energy transfer type, the carriers injected from the two electrodes recombine within the host layer and the host material is excited. Energy transfer occurs from the excited host material to the dopant material, and finally emission is obtained from the dopant material. In the carrier trap type, carriers which have moved through the host layer directly recombine on the dopant material, and the excited dopant emits light. In each case, if there is used as the dopant material, which has the emissive function, a material with high chromatic purity in the solution state and a high photoluminescent quantum yield, it is possible to obtain high chromatic purity and high luminance efficiency. Furthermore, the addition of a dopant material serves to lower the crystallinity of the host layer film, which is the film parent body, and for this reason too the durability is enhanced.

**[0018]** In the case where there is used this kind of combination of host material and dopant material, the dopant material can be contained within the entirety of the host material or it may be contained in a part thereof. Furthermore, the dopant material may be provided as a layer or it may be dispersed.

**[0019]** The organic material in the case where it alone forms the emissive layer, or the host material in the case of a combination of host and dopant materials, may be a derivative of a condensed ring system such as anthracene, pyrene or perylene, a derivative of a heterocycle such as pyrazine, naphthyridine, quinoxaline, pyrrolopyridine, pyrimidine, thiophene or thioxanthene, a quinolinol-metal complex such as the tris(8-quinolinolato)aluminium complex, a benzoquinolinol-metal complex, a bipyridine-metal complex, a rhodamine-metal complex, an azomethine-metal complex, a distyrylbenzene derivative, a tetraphenylbutadiene derivative, a stilbene derivative, an aldazine derivative, a coumarin derivative, a phthalimide derivative, a naphthalimide derivative, a perinone derivative, a pyrrolopyrrole derivative, a cyclopentadiene derivative, an imidazole derivative, an oxazole derivative, a thiazole derivative, an oxadiazole derivative, a thiadiazole derivative, a triazole derivative or other such azole derivative or metal complex thereof, a benzoxazole, benzimidazole, benzothiazole or other such benzazole derivative or metal complex thereof, an amine derivative such as a triphenylamine derivative or carbazole derivative, a merocyanine derivative, a porphyrin derivative, tris(2-phenylpyridine)iridium complex or other such phosphorescent material or, in the case of polymer systems, a polyphenylene vinylene derivative, poly-p-phenylene derivative or polythiophene derivative, or the like.

**[0020]** Examples of the dopant material are anthracene, perylene and other such condensed polycyclic aromatic hydrocarbons, 7-dimethylamino-4-methylcoumarin and other such coumarin derivatives, bis(diisopropylphenyl)-perylenetetracarboxylic imide and other such naphthalimide derivatives, perinone derivatives, rare earth complexes such as Eu complexes with an acetylacetone, benzoylacetone or phenanthroline ligand, dicyanomethylene pyran derivatives, dicyanomethylene thiopyran derivatives, magnesium phthalocyanine, aluminium chlorophthalocyanine and other such metal-phthalocyanine derivatives, porphyrin derivatives, rhodamine derivatives, deazaflavin derivatives, coumarin derivatives, oxazine compounds, thioxanthene derivatives, cyanine dye derivatives, fluorescein derivatives, acridine derivatives, quinacridone derivatives, pyrrolopyrrole derivatives, quinazoline derivatives, pyrrolopyridine derivatives, squarilium derivatives, violanthrone derivatives, phenazine derivatives, acridone derivatives, deazaflavin derivatives, pyrromethene derivatives and their metal complexes, phenoxazine derivatives, phenoxazone derivatives, thiadiazolopyrene derivatives, tris(2-phenylpyridine)iridium complex, tris(2-phenylpyridyl)-iridium complex, tris[2-(2-thiophenyl)pyridyl]iridium complex, tris[2-(2-benzothiophenyl)pyridyl]iridium complex, tris(2-phenylbenzothiazol)iridium complex, tris(2-phenylbenzoxazole)iridium complex, trisbenzoquinoline-iridium complex, bis(2-phenylpyridyl)(acetylacetonato)iridium complex, bis(2-(2-thiophenyl)pyridyl]iridium complex, bis[2-(2-benzothiophenyl)pyridyl](acetylacetonato)iridium complex, bis (2-phenylbenzothiazole)(acetylacetonato)-iridium complex, bis(2-phenylbenzoxazole)(acetyl-acetonato)iridium complex, bisbenzoquinoline(acetyl-acetonato)iridium complex, platinum-porphyrin complex and other such phosphorescent materials. These may be used on their own or a mixture of a plurality of such derivatives may be used.

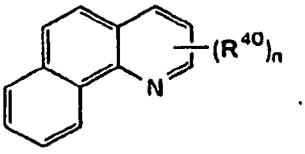
**[0021]** Furthermore, for the purposes of modifying the film properties or trapping excess carriers and enhancing the durability, there may sometimes be added a dopant material without it having an emissive capacity. As the dopant material in such circumstances, there is selected from amongst various organic and inorganic compounds one which corresponds to the host material. The doping conditions are the same as above.

**[0022]** In terms of the objective of achieving efficient recombination of the holes and the electrons within the emissive layer, it is preferred that the light emitting device of the present invention also has a hole transporting layer between the anode and the emissive layer. The hole transporting layer is a layer which further transports the holes after they have been injected from the anode. Examples of hole transporting materials are N,N'-diphenyl-N,N'-bis(3-methylphenyl)-4,4'-diphenyl-1,1'-diamine, N,N'-bis(1-naphthyl)-N,N'-diphenyl-4,4'-diphenyl-1,1'-diamine and other such triphenylamines, bis(N-arylcarbazoles) or bis(N-alkylcarbazoles), pyrazoline derivatives, stilbene derivatives, distyryl derivatives, hydrazone compounds, oxadiazole derivatives, phthalocyanine derivatives, porphyrin derivatives and other such heterocyclic compounds and, in the case of polymer systems, polycarbonates or styrene derivatives with the aforesaid monomers in side chains, polyvinylcarbazole, polysilanes and the like. These may be used on their own or there may be used a plurality in the form of a mixture or multilayer. Furthermore, for the purposes of enhancing the transportation capacity of the entire hole transporting layer, or for enhancing the thermal stability or electrochemical stability, the formation of the hole transporting layer may be carried out with the addition of an organic compound, inorganic compound or metal complex which does not have a hole transporting capacity.

**[0023]** The anode in the present invention should be transparent in order to extract the light. Examples include electroconductive metal oxides such as tin oxide, indium oxide and indium tin oxide (ITO), or metals such as gold, silver and chromium, inorganic electroconductive substances such as copper iodide and copper sulphide, and electroconductive polymers such as polythiophene, polypyrrole and polyaniline. The use of ITO glass or NESA glass is particularly preferred. With regard to the resistance of the transparent electrode, it should be possible to supply sufficient current to achieve emission and a low resistance is preferred from the point of view of the power consumption of the device. For example, an ITO substrate of resistance no more than 300 Ω/□ will function as a device electrode but, since there currently exist substrates of resistance value about 10 Ω/□, using a low resistance product is particularly preferred. The thickness of the ITO can be freely selected in conjunction with the resistance value but normally there is usually employed a thickness in the range 100-300 nm. Furthermore, soda-lime glass, alkali-free glass or the like may be used as the glass substrate. Its thickness should be at least 0.5 mm in order to ensure mechanical strength. With regard to the type of glass material, an alkali-free glass is preferred in that there is little ion-elution from the glass but soda-lime glass can also be used where it has been given a barrier coating of $SiO_2$ or the like. Moreover, providing that the anode functions stably, the anode may also be formed on a plastic substrate. As examples of methods for forming an ITO film, there are the electron beam method, the sputtering method and the chemical reaction method, etc.

**[0024]** The cathode may be any substance which can efficiently inject electrons into the organic layer. Examples of the cathode material are platinum, gold, silver, copper, iron, tin, zinc, aluminium, indium, chromium, lithium, sodium, potassium, calcium, magnesium, caesium, strontium and the like. For the purposes of raising the electron injection efficiency and enhancing the characteristics of the device, lithium, sodium, potassium, calcium, magnesium, caesium, strontium or alloys of such low work function metals are effective. Furthermore, the method of doping the organic layer with a small amount of lithium, magnesium or caesium (a vacuum vapour-deposited film of no more than 1 nm as determined by means of a film thickness monitor) and using an electrode of high stability is also preferred, and moreover it is possible to employ an inorganic salt such as lithium fluoride. Again, for electrode protection, it is preferred that lamination be carried out with a metal such as platinum, gold, silver, copper, iron, tin, aluminium, indium· or alloy of such metal, an inorganic material such as silica, titania or silicon nitride, or polyvinyl alcohol, vinyl chloride or a hydrocarbon polymer, etc. Examples of methods for the production of such electrodes are the resistance heating, electron beam, sputtering, ion plating and coating methods, etc.

**[0025]** The benzoquinoline skeletal structure in the present invention is a skeletal structure comprising benzene condensed to quinoline at any position, and it is represented by general formulae (9) to (14).



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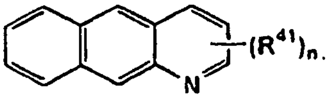

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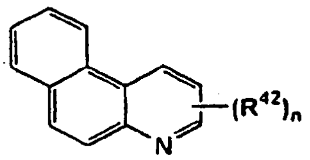

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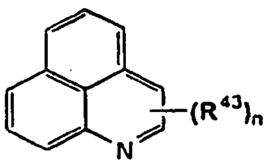

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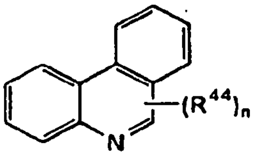
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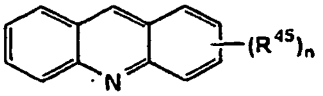
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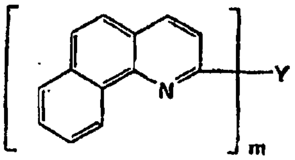
**[0026]** $R^{40}$ to $R^{45}$ in general formulae (9) to (14) denote substituent groups at any position in the benzoquinoline skeletal structure other than a position used for connection to another benzoquinoline skeletal structure, and are selected from amongst hydrogen, alkyl groups, cycloakyl groups, aralkyl groups, alkenyl groups, cycloalkenyl groups, alkoxy groups, alkylthio groups where the oxygen atom of the ether linkage in an alkoxy group is replaced by a sulphur atom, aryl ether groups, aryl thioether groups where the oxygen atom of the ether linkage in an aryl ether group is replaced by a sulphur atom, aryl groups, heterocyclic groups, halogens, cyano group, aldehyde group, carbonyl group, carboxyl group, ester group, carbamoyl group, amino group, nitro group, silyl groups, siloxanyl groups which are groups with silicon having an interposed ether linkage, and cyclic structures formed with an adjacent substituent group, n is an integer in the range 1 to 8 and, in the case where there are a plurality of substituents, said substituents may be the same or different.

**[0027]** Furthermore, in order to obtain stable emission over a long period, a material with excellent thermal stability and thin-film forming properties is desirable.

**[0028]** In order to obtain high luminance, the use of a compound with a high electron transporting capacity is preferred. Hence, as examples of the aforesaid compounds with a plurality of benzoquinoline skeletal structures, those compounds where said plurality of benzoquinoline skeletal structures are connected by a conjugated bond, aromatic hydrocarbon, aromatic heterocycle or mixture of these.

**[0029]** Taking the case where the basic skeletal structure is represented by general formula (9), specific examples of compounds based on the benzoquinoline parent skeletal structure are those with a structure represented by general formula (15).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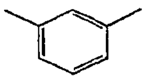
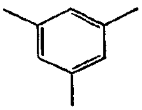
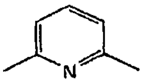
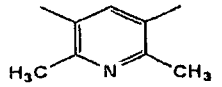
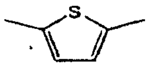
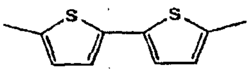
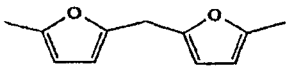
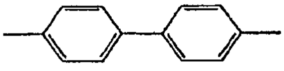
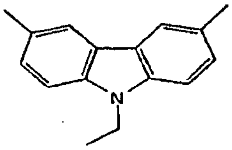
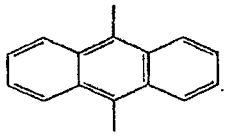
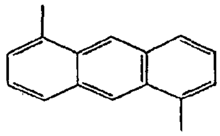
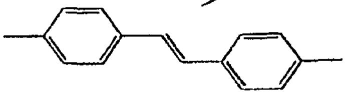
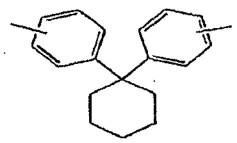
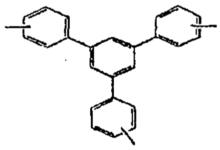
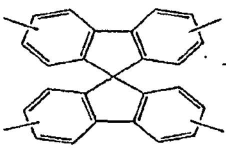
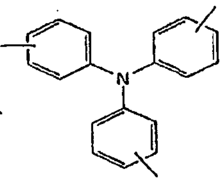
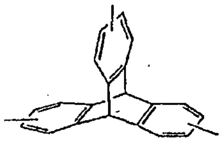
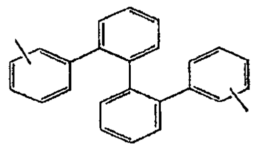
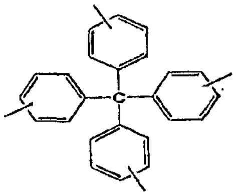
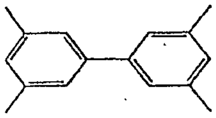
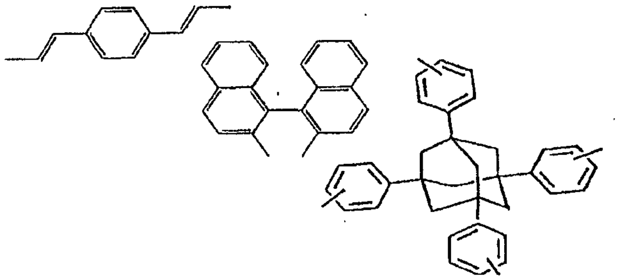
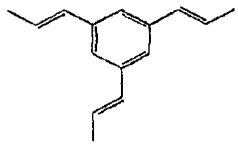
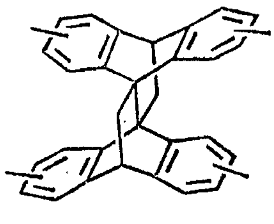
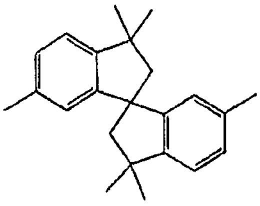
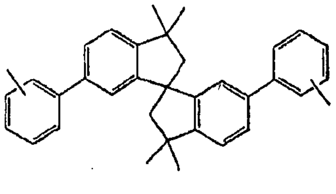
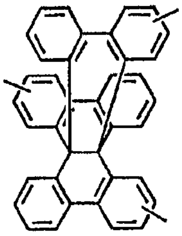
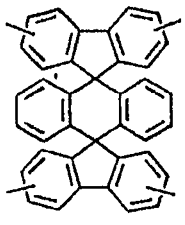
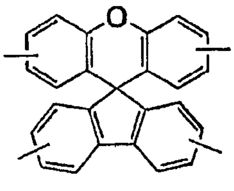
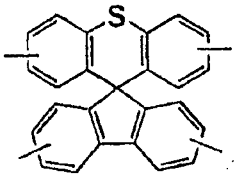
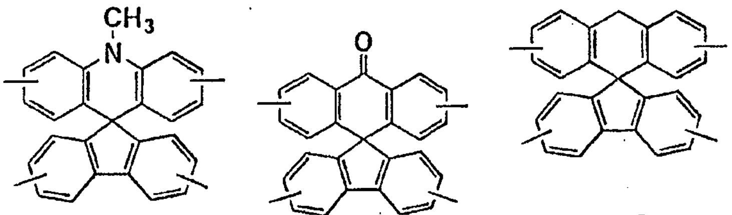
**[0030]** Here, Y is a correcting unit which contrains a conjugated bond or conjugated bands, an aromatic hydrocarbon, an aromatic heterocycle or mixture thereof, and m is 2 or more.

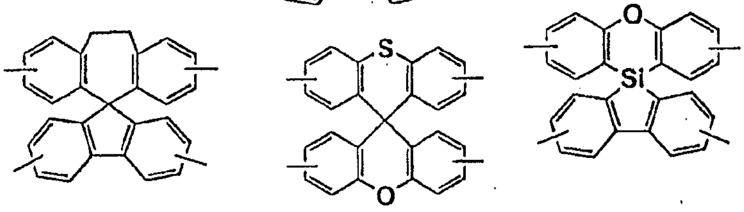
**[0031]** With regard to the connecting units for connecting together the benzoquinoline structures, connecting units which contain conjugated bonds, aromatic hydrocarbons or aromatic heterocycles are used, and the following can be given as specific examples.

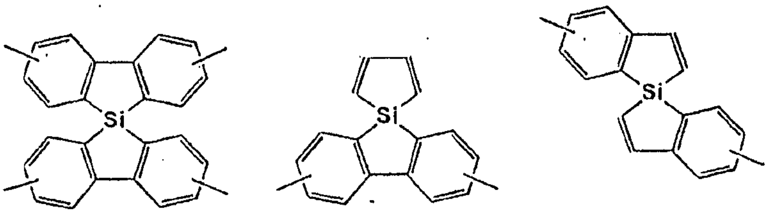


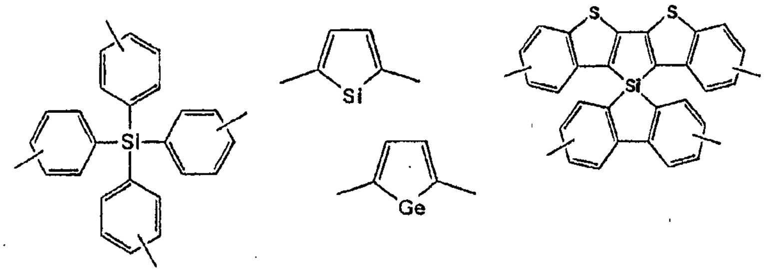






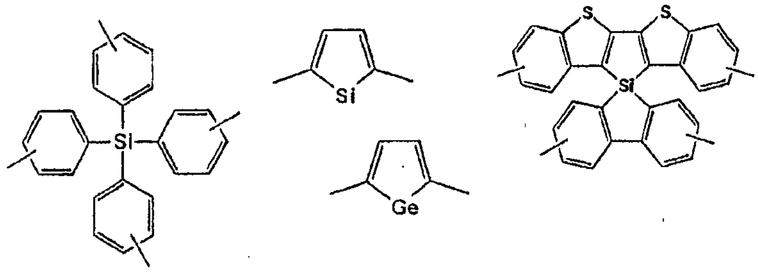



































**[0032]** There may be used one type of connecting unit or a mixture thereof.

**[0033]** These connecting units can be obtained commercially or they can be synthesized in accordance with normal procedures. Specific examples for a number of skeletal structures are shown below.

**[0034]** The synthesis of the 9,9'-spirobifluorene structure is described, for example, in J. Am. Chem. Soc., vol.52 (1930), page 2881, and in US Patent 5840217 in the section "EXAMPLES A. Starting compounds a) Synthesis of 9,9'-spirobifluorene". Thus, 2-bromobiphenyl is converted to the Grignard in THF using metal magnesium, then reacted with 9-fluorenone at room temperature to 50°C and treatment carried out in the usual way, after which the hydroxy compound obtained is subjected to heating and elimination of water in acetic acid to which a small amount of hydrochloric acid has been added and treatment performed in the usual way.

**[0035]** The synthesis of the 9,9'-spirobi(9H-9-silafluorene) structure is described in J. Am. Chem. Soc., von. 80 (1958), page 1883. This can be obtained by reacting 2,2'-dibromobiphenyl with metal lithium in ether, and then performing reaction with tetrachlorosilane at a specified temperature, followed by treatment in the usual way.

**[0036]** The synthesis of the hexabenzopropellane structure is described in for example Libigs Ann. Chem., vol.749 (1971) page 38. 9-fluorenone is reacted with triethyl phosphite, then treatment performed with methanol and the spiroketone compound obtained. Next, reaction is conducted in ether at a specified temperature between a 2-bromobiphenyl lithio compound and the spiroketone compound, followed by treatment in the usual way, and then hydroxy compound obtained is subjected to heating and elimination of water in acetic acid with the addition of methanesulphonic acid, and treatment performed in the usual way.

**[0037]** The benzoquirioline derivatives of the present invention are outstanding in their electron transporting capacity and hole blocking capacity, so are preferably used as the electron transporting material.

**[0038]** The compounds of the present invention which possess a plurality of benzoquinoline skeletal structures can be synthesized by the same kinds of methods as phenanthroline derivatives. After introducing acetyl groups into the connecting unit, reaction is performed with a naphthalene derivative to form the benzoquinoline rings (literature reference: J. Org. Chem. 1996, 61, page 3021 "1,3-Di(benzo([h]quinolin-2-yl)benzene") or, alternatively, after introducing reactive substituents such as iodo or bromo groups, addition of the benzoquinoline rings is effected.

**[0039]** Examples of methods for forming the respective layers are evaporation by resistance heating, electron beam evaporation, sputtering, molecular deposition, coating and the like. Normally, evaporation by resistance heating and electron beam evaporation are preferred in terms of properties. Layer thickness will depend on the resistance of the emissive substance, so cannot be restricted but will be selected from within the range 1 to 1,000 nm.

**[0040]** Reference to electrical energy primarily means direct current but it is also possible to use a pulse current or alternating current. The values of the current and voltage are not particularly restricted but, taking into account power consumption and the life of the device, the maximum luminance should be obtained at as low energy as possible.

**[0041]** Reference to matrix in the present invention means the matrix array of pixels used for display, and by association of pixels the display of characters or images is effected. The shape and size of the pixels is determined by the application. In the case of image and character display by personal computers, monitors and televisions, there are normally used square-shaped pixels with up to 300 μm sides, and in the case of large-size displays such as display panels there are normally used pixels with sides of the mm order. In the case of a monochrome display, there may be arrayed pixels of the same colour but, in the case of a colour display, red, green and blue pixels are arranged side by side. In such circumstances, typically there are delta and stripe types. The method of driving the matrix may be either the active matrix or passive matrix driving method. Construction is simpler in the case of passive matrix driving, while an active matrix may be superior in operational characteristics, so here too selection will be made according to the application.

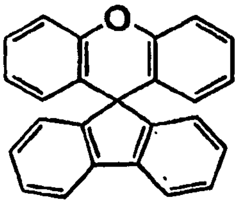
**[0042]** Segment type in the present invention means that a pattern is formed so as to display previously-determined data, and there is emission in a predetermined region. Examples include time and temperature displays by digital watches and thermometers, operating-state displays in the case of audio equipment and microwave ovens, vehicle panel displays and the like. Now, the aforesaid matrix and segment displays may also both be present in the same panel.

**[0043]** The light emitting device of the present invention can also be favourably employed as a back light. A back light is primarily used for the purposes of enhancing the visibility of a display means which is not self-illuminating, and it may be employed for liquid crystal display devices, watches, audio equipment, automobile panels, signboards, signs and the like. In particular, liquid crystal display devices and, especially, conventional personal computers, have comprised fluorescent bulbs or light-guiding sheets, so making these thinner has been difficult. However, thin, lightweight, products are possible with backlights employing the light emitting device of the present invention.

**[0044]** Below, the present invention is explained by providing examples and comparative examples. Among these, examples 1-3, 6-22, 34-51 and 53-54 are not according to the present invention, but are useful for understanding the same. The conditions employed and the results obtained are shown together in Table 1 and Table 2 at the end of this section.

Example 1 (Synthesis of Connecting Unit 1)

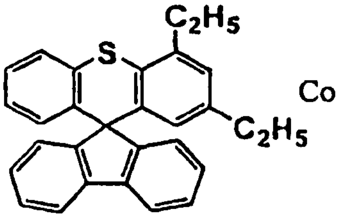
**[0045]** 14.8 g of 2-bromobiphenyl was converted to the Grignard form in THF using 2.2 g of metal magnesium, then this reacted with 12.3 g of 9-xanthone at from room temperature to 50°C and, by treatment in the normal way, 9-(2-biphenyl)-9-xanthenol was obtained. This was heated in acetic acid to which a small amount of hydrochloric acid had been added, so that water was eliminated and, by treatment in the usual way, Connecting Unit 1 (8.5 g) shown below was obtained. $^1$H-NMR (CDCl$_3$, ppm): 7.8 (d, 2H), 7.2 (t, 2H), 7.2 (m, 8H), 6.8 (t, 2H), 6.4 (d, 2H)



Connecting Unit 1

Example 2 (Synthesis of Connecting Unit 2)

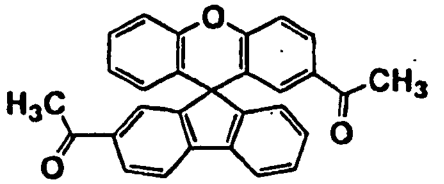
**[0046]** 11.9 g of 2-bromobiphenyl was converted to the Grignard form in THF using 1.7 g of metal magnesium, then this reacted with 13.4 g of 2,4-diethyl-9-thioxanthone at from room temperature to 50°C and, by treatment in the normal way, 2,4-diethyl-9-(2-biphenyl)-9-thioxanthenol was obtained. This was heated in acetic acid to which a small amount of hydrochloric acid had been added, so that water was eliminated and, by treatment in the usual way, Connecting Unit 2 (13.8 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm) : 7.8 (m, 2H), 7.6 (d, 2H), 7.4 (m, 3H), 7.2 (m, 2H), 7.1 (t, 1H), 6.9 (s, 1H), 6.8 (t, 1H), 6.5 (d, 1H), 6.2 (s, 1H), 2.9 (m, 2H), 2.3 (m, 2H), 1.4 (t, 3H), 0.9 (t, 3H)



Connecting Unit 2

Example 3 (Introduction of acetyl groups into Connecting Unit 1: Connecting Unit 1')

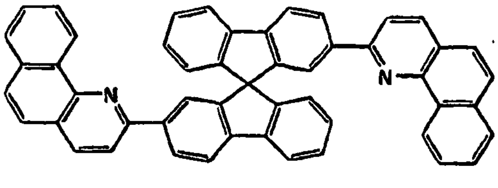
**[0047]** Connecting Unit 1 (8.5 g) was reacted with 4.5 g of acetyl chloride and 7.5 g of aluminium chloride at 56°C in 1,2-dichloroethane and, by treatment in the normal way, Connecting Unit 1' (13.1 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm) : 8.0 (d, 1H), 7.9 (d, 2H), 7.8 (d, 1H), 7.7 (s, 1H), 7.4 (t, 1H), 7.3-7.1 (m, 5H), 7.0 (s, 1H), 6.8 (t, 1H), 6.3 (d, 1H), 2.5 (s, 3H), 2.3 (s, 3H)



Connecting Unit 1'

Example 4 (Synthesis of the benzoquinoline derivative (BQ-1))

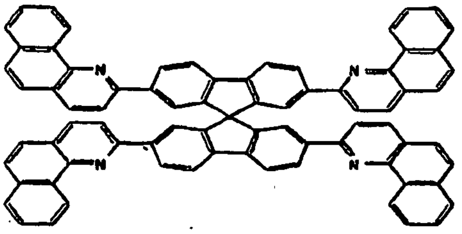
**[0048]** 23.8 g of 2-bromobiphenyl was converted to the Grignard form in THF using.3.4 g of metal magnesium, then this reacted with 18.0 g of 9-fluorenone at from room temperature to 50°C and, by treatment in the normal way, 9-(2-biphenyl)-9-fluorenol was obtained. This was heated in acetic acid to which a small amount of hydrochloric acid had been added, so that water was eliminated and, by treatment in the usual way, Connecting Unit 1 (18.5 g) was obtained. Next, Connecting Unit 1 (15.8 g) was reacted with 8.6 g of acetyl chloride and 14.7 g of aluminium chloride at 50°C in 1,2-dichloroethane and, by treatment in the normal way, 2,2'-diacetyl-9,9'-spirobifluorene (11.2 g) was obtained. This diacetyl compound (2.3 g) was reacted with 2.0 g of 1-amino-2-naphthalenecarbaldehyde and 1.6 g of potassium hydroxide at 60°C in dioxane and, by treatment in the normal way, there was obtained BQ-1 (1.5 g) shown below. $^1$H-NMR ($CDCl_3$, ppm) : 9. 4 (d, 2H), 8.6 (d·d, 2H), 8.1 (d, 2H), 8.0 (t, 4H), 7.8 (d, 2H), 7.8-7.6 (m, 12H), 7.4 (t, 2H), 7.2 (t, 2H), 6.8 (d, 2H)



BQ-1

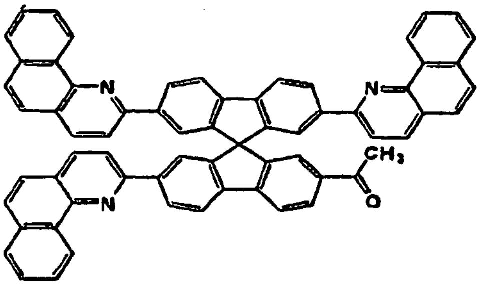
Example 5 (Synthesis of benzoquinoline derivatives (BQ-2 and 3))

**[0049]** Reaction was carried out in the same way as in Example 4 using the tetra-acetyl derivative, and treatment was carried out in the normal way so that BQ-2 and BQ-3 shown below were obtained.



**BQ-2**

BQ-2: $^1$H-NMR ($CDCl_3$, ppm) : 9.39 (d, 4H), 8.72 (d, 4H), 8.27 (d, 4H), 8.03 (d, 4H), 7.84-7.55 (m, 24H), 7.32 (d·d·d, 4H)

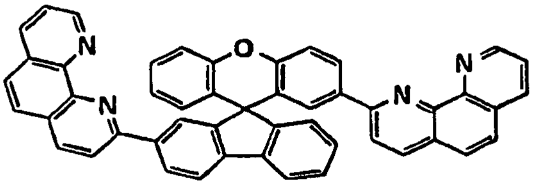


**BQ-3**

BQ-3: $^1$H-NMR ($CDCl_3$, ppm) : 9.36 (d, 3H), 8.72 (d·d, 1H), 8.66 (d·d, 2H), 8.24 (d,1H), 8.23 (d, 2H), 8.12-8.03 (m, 6H), 7.87-7.49 (m, 21H), 2.46 (s, 3H)

Example 6 (Synthesis of the phenanthroline derivative (Phen-1))

**[0050]** Connecting Unit 1' (5.0 g) was reacted with 5.2 g of 8-amino-7-quinolinecarbaldehyde and 5.0 g of potassium hydroxide at 60°C in dioxane and, by treatment in the normal way, Phen-1 (5.8 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm): 9.2 (d, 2H), 8.8 (d, 1H), 8.5 (d, 1H), 8.2 (m, 3H), 8. 1 (t, 2H), 7.9 (t, 2H), 7.7-7.5 (m, 8H), 7.4 (m, 1H), 7.3-7.2 (m, 5H), 7.1 (s, 1H), 6.8 (t, 1H), 6.5 (d, 1H)

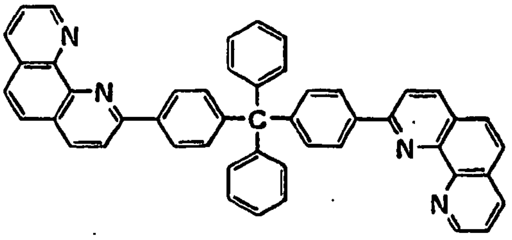


**Phen-1**

Example 7 (Synthesis of phenanthroline derivative (Phen-2))

**[0051]** 2.5 g of tetraphenylmethane, 2.08 g of aluminium chloride and 1.22 ml of acetyl chloride were added to 100 ml of 1,2-dichloroethane, and reaction carried out for 1.5 hours at room temperature and then for 2 hours at 70°C. By treatment in the normal way, 1.03 g of the diacetyltetraphenylmethane was obtained. $^1$H-NMR ($CDCl_3$, ppm) : 7.86 (d, 4H), 7.35 (d, 4H), 7.24 (m, 10H)

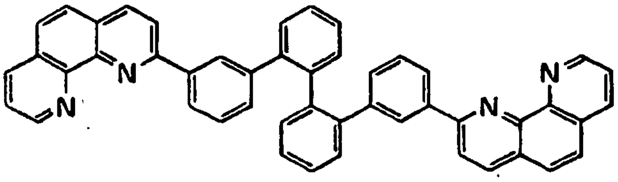
**[0052]** 1.0 g of the aforesaid diacetyltetraphenylmethane was reacted with 1.06 g of 8-amino-7-quinolinecarbaldehyde and 1.0 g of potassium hydroxide in dioxane at 60°C and, by treatment in the normal way, Phen-2 (1.21 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm): 9.21 (d·d, 2H), 8.31-8.19 (m, 8H), 8.07 (d, 2H), 7.76 (q, 4H), 7.62 (d·d, 2H), 7.50 (d, 4H), 7.39-7.20 (m, 10H)



Phen-2

Example 8 (Synthesis of the phenanthroline derivative (Phen-3))

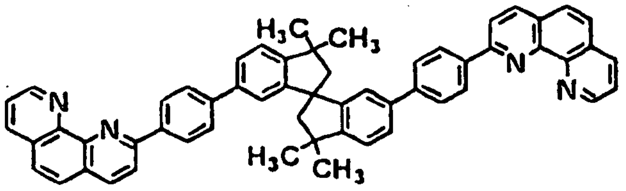
**[0053]** 2.5 g of 2,2'-dibromobiphenyl, 3.9 g of 3-acetylphenyl-boronic acid, 21 ml of 2M sodium carbonate and 0.37 g of tetrakis(triphenylphosphine)palladium(0) were added to 200 ml of 1,2-dimethoxyethane, and refluxing carried out for 10 hours under nitrogen so that a Suzuki coupling reaction was performed and, by treatment in the normal way, 0.57 g of 2,2'-bis(3-acetylphenyl)biphenyl was obtained. 0.57 g of this diacetyl derivative was reacted with 0.63 g of 8-amino-7-quinolinecarbaldehyde and 0.6 g of potassium hydroxide in dioxane at 60°C and, by treatment in the normal way, Phen-3 (0.76 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm): 9.20 (dd, 2H), 8.43 (d, 2H), 8.16 (d·d, 2H), 7.79 (d, 2H), 7.61-7.26 (m, 18H), 7.17 (t, 2H), 6.77 (d, 2H)



Phen-3

Example 9 (Synthesis of the phenanthroline derivative (Phen-4))

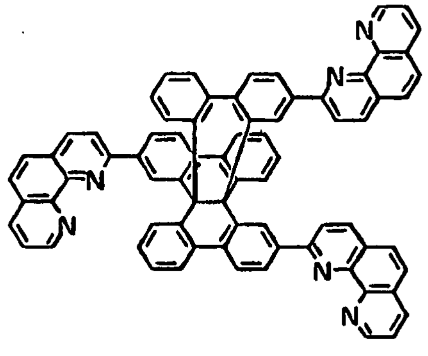
**[0054]** 2.5 g of 6,6'-bis(trifluoromethanesulphonyloxy)-3,3,3',3'-tetramethyl-1,1'-spirobiindane, 2.2 g of 4-acetylphenylboronic acid, 12 ml of 2M sodium carbonate and 0.40 g of tetrakis(triphenylphosphine)palladium(0) were added to a solvent mixture of 50 ml of toluene and 30 ml of ethanol, and then refluxing carried out for 12 hours under nitrogen so that a Suzuki coupling reaction was conducted and, by treatment in the normal way, 1.5 g of 6,6'-bis(4-acetylphenyl)-3,3,3',3'-tetrarnethyl-1,1'-spirobiindane was obtained. 1.50 g of this diacetyl derivative was reacted with 1.26 g of 8-amino-7-quinolinecarbaldehyde and 0.9 g of potassium hydroxide in dioxane at 60°C and, by treatment in the normal way, Phen-4 (1.40 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm): 9.21 (d, 2H), 8.35 (d, 4H), 8.25 (t, 4H), 8.09 (d, 2H), 7.80-7.59 (m, 12H), 7.34 (d, 2H), 7.24 (d, 2H), 2.46 (d, 4H), 1.52 (s, 6H), 1.46 (s, 6H)



Phen-4

Example 10 (Synthesis of the phenanthroline derivative (Phen-5))

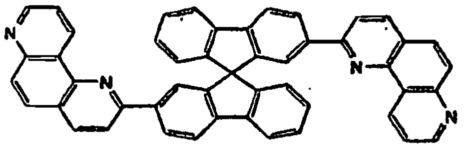
**[0055]** 4.0 g of propellane, 3.3 g of aluminium chloride and 2.0 ml of acetyl chloride were added to 200 ml of 1,2-dichloroethane, and reaction carried out for 1.5 hours at room temperature and then for 6 hours at 60°C. By treatment in the normal way, 3.26 g of the triacetyl-propellane was obtained. 1.41 g of this triacetyl derivative was reacted with 1.50 g of 8-amino-7-quinolinecarbaldehyde and 1.5 g of potassium hydroxide in dioxane at 60°C and, by treatment in the normal way, Phen-5 (1.32 g) shown below was obtained.



**Phen-5**

Example 11 (Synthesis of the phenanthroline derivative (Phen-6))

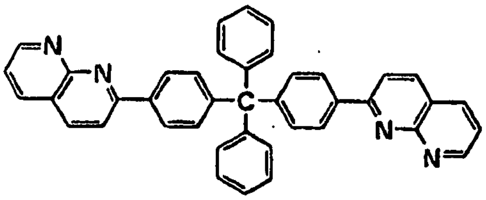
**[0056]** 1.52 g of 2,2'-diacetyl-9,9'-spirobifluorene was reacted with 1.31 g of 5-amino-6-quinolinecarbaldehyde and 1.0 g of potassium hydroxide in dioxane at 60°C and, by treatment in the normal way, Phen-6 (0.29 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm) : 9.57 (d, 2H), 8.99 (d·d, 2H), 8.57 (d·d, 2H), 8.11 (d·d, 4H), 7.98 (t, 4H), 7.84 (d, 2H), 7.81 (d, 2H), 7.61-7.56 (m, 4H), 7.45 (t, 2H), 7.18 (t, 2H), 6.84 (d, 2H).



**Phen-6**

Example 12 (Synthesis of the naphthyridine derivative (TPM-dNTR))

**[0057]** 0.77 g of diacetyltetraphenylmethane obtained by the method described in Example 7 was reacted with 0.51 g of 2-aminonicotinaldehyde and 0.76 g of potassium hydroxide in dioxane and, by treatment in the normal way, TPM-dNTR (0.82 g) shown below was obtained. $^1$H-NMR ($CDCl_3$, ppm): 9.12 (d·d, 2H), 8.27-8.16 (m, 8H), 8. 00 (d, 2H), 7.49-7.44 (m, 6H), 7.34-7.23 (m, 10H)

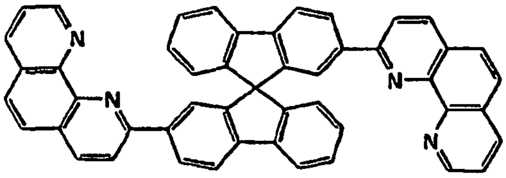


**TPM-dNTR**

Example 13

**[0058]** A glass substrate on which a 150 nm ITO transparent electroconductive film had been deposited (produced by the Asahi Glass Co., 15 Ω/□, electron beam vapour-deposited product) was cut to 30 x 40 mm and etching carried out. The substrate obtained was subjected to ultrasonic washing for 15 minute periods with acetone and "Semico-Clean 56" respectively, after which washing was carried out with ultrapure water. Next, 15 minutes ultrasonic washing was performed with isopropyl alcohol, after which it was immersed for 15 minutes in hot methanol and dried. Just prior to producing the device, this substrate was given a 1 hour UV/ozone treatment, then placed in vacuum vapour-deposition equipment and the equipment evacuated until the degree of vacuum inside was less than 1 x $10^{-5}$ Pa. By means of a resistance heating method, firstly there was deposited 10 nm of copper phthalocyanine (CuPc) as a first hole injecting and transporting layer, and then there was provided a 50 nm layer of N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1'-diphenyl-4,4'-diamine (α-NPD) as a second hole transporting layer. Furthermore, next there was codeposited to a thickness of 25 nm the emissive layer region using tris(8-quinolinolato)aluminium (III) (Alq3) as the host material and 2,3,5,6-tetrahydro-9-(2-benzothiazolyl)-quinolizinb-[9,9a,1-gh]coumarin (Coumarin-1) as the dopant material, such that there was 1.0 wt% of the dopant. Thereafter, ETM1 shown below was deposited at a thickness of 25 nm as the electron transporting layer. Next, 0.2 nm lithium doping was performed, and finally there was vapour-deposited 150 nm of aluminium to produce the cathode and, in this way, a 5 x 5 mm square device was produced. The ionization potential of the electron transporting layer was 6.07

eV, the molecular weight was 672, the glass transition temperature was 219°C and the cold crystallization temperature was 352°C. The ionization potential of the emissive layer was 5.78 eV and so the difference in the ionization potential compared to the emissive layer was 0.29 eV. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the dopant material of peak emission wavelength 523 nm, and the luminance was 38,000 cd/m$^2$. Furthermore, after the elapse of 500 hours operation of this light emitting device, the percentage retention of the initial luminance was 80% and a uniform emissive surface was maintained.



ETM1

Comparative Example 1

**[0059]** A light emitting device was constructed in exactly the same way as in Example 13 except that there was used Alq3 as the electron transporting layer. The ionization potential of Alq3 is 5.79, its molecular weight is 459 and its glass transition temperature is 180°C. From this light emitting device, at a driving voltage of 10 V there was obtained green coloured light emission based on the dopant material of peak emission wavelength 523 nm, and the luminance was 6000 cd/m$^2$.

Comparative Example 2

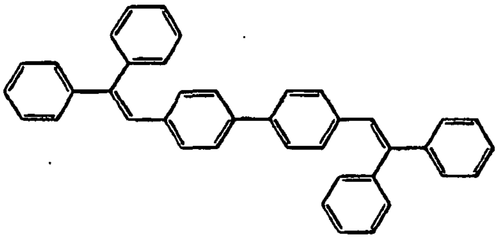
**[0060]** A light emitting device was constructed in exactly the same way as in Example 13 except that there was used 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) as the electron transporting layer. The ionization potential of BCP is 6.2 eV, its molecular weight is 360 and its crystallization temperature is 77°C. From this light emitting device, at a driving voltage of 10 V there was obtained green coloured light emission based on the dopant material of peak emission wavelength 523 nm, and the luminance was 12,000 cd/m$^2$. However, after the elapse of 500 hours operation of this light emitting device, the percentage retention of the initial luminance was less than 50% and unevenness was apparent at the emissive surface.

Example 14

**[0061]** A light emitting device was constructed in exactly the same way as in Example 13 except that there was deposited, as the emissive layer region, a 20 nm thickness of a mixture of 4,4'-bis(N-carbazolyl)biphenyl (CBP) and tris (2-phenylpyridine)iridium complex (Ir(ppy)3) [iridium complex content = 8 wt%]. At a driving voltage of 6 V, from this light emitting device there was obtained green coloured light emission based on the iridium complex of peak emission wavelength 515 nm, and the luminance was 1000 cd/m$^2$. Thus, the electron transporting material of the present invention also functioned effectively in a device employing a phosphorescent material as an emissive material.

Example 15

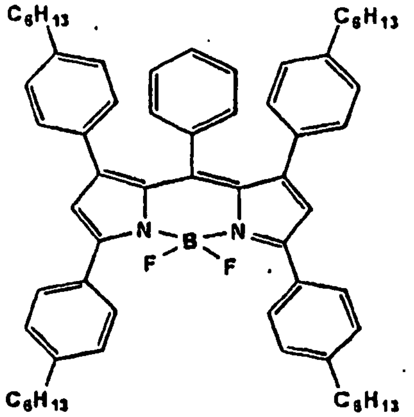
**[0062]** A light emitting device was constructed in exactly the same way as in Example 13 except that there was used EM1, which is shown below, as the emissive material. At a driving voltage of 15 V, from this light emitting device there was obtained blue coloured light emission based on the EM1 of peak emission wavelength 463 nm, and the luminance was 8,000 cd/m$^2$. Thus, the electron transporting material of the present invention also functioned effectively in a blue coloured light emitting device.



EM1

Example 16

**[0063]** A light emitting device was constructed in exactly the same way as in Example 13 except that as the emissive layer region co-deposition was carried out at a thickness of 15 nm using 1,4-diketo-2,5-bis(3,5-dimethylbenzyl)-3,6-bis (4-methylphenyl)pyrrolo[3,4-c]pyrrole (DPP-1) as the host material and EM2 shown below as the dopant material, such that there was 1.0 wt% of the dopant.



EM2

Comparative Example 3

**[0064]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used Alq3 as the electron transporting layer.

Example 17

**[0065]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used the Phen-3 shown in Example 8 as the electron transporting layer. The ionization potential of the electron transporting layer was 6.14 eV, the molecular weight was 663 and the glass transition temperature was 150°C.

Example 18

**[0066]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used 1,4-diketo-2,5-bis(4-methylbenzyl)-3,6-bis(1-naphthyl)pyrrolo[3,4-c]pyrrole (DPP-2) as the host material.

Example 19

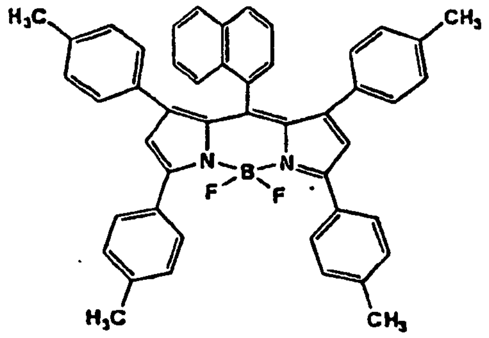
**[0067]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used 1,4-diketo-2,5-dibenzyl-3,6-bis(1-naphthyl)pyrrolo[3,4-c]pyrrole (DPP-3) as the host material.

Example 20

**[0068]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used 1,4-diketo-2,5-bis(4-isopropylbenzyl)-3,6-bis(1-naphthyl)pyrrolo[3,4-c]pyrrole (DPP-4) as the host material.

Example 21

**[0069]** A light emitting device was prepared in exactly the same way as in Example 16 except that there was used 1,4-diketo-2,5-diethyl-3,6-bis(1-naphthyl)pyrrolo[3,4-c]pyrrole (DPP-5) as the host material and EM3 shown below as the dopant material.



EM3

Example 22

**[0070]** A light emitting device was prepared in exactly the same way as in Example 21 except that there was used 1,4-diketo-2,5-bis(4-methylbenzyl)-3,6-bis(1-(4-tolyl)naphthyl)pyrrolo[3,4-c]pyrrole (DPP-6) as the host material.

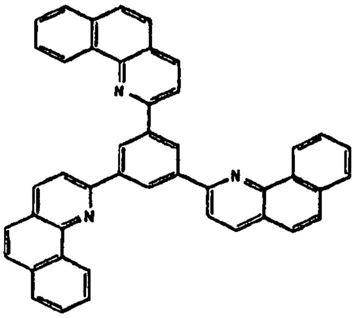
**[0071]** Table 1 shows the results obtained in the case of Examples 16 to 22 where the electron transporting material of the present invention was applied to a red coloured light emitting device. By employing the electron transporting material of the present invention, red light emission of high chromatic purity and high luminance was obtained.

Example 23

**[0072]** A light emitting device was constructed in exactly the same way as in Example 13 except that for the emissive layer region there was employed the BQ-1 of Example 4 as the host material and 4,4-difluoro-1,3,5,7,8-tetramethyl-4-bora-3a,4a-diazaindacene (PM-1) as the dopant material such that the dopant concentration was 0.5%, and for the electron transporting layer there was used BQ-1 in the same way. At a driving voltage of 11 V, from this light emitting device there was obtained green coloured light emission based on the dopant peak emission wavelength of 519 nm, and the luminance was 8,000 cd/m$^2$.

Example 24

**[0073]** The same procedure was followed as in Example 13 up to the provision of the hole transporting material layer. Next, as the emissive material there was provided Alq3 at a thickness of 15 nm, and then as the electron transporting material there was provided a layer of ETM2, which is shown below, at a thickness of 35 nm. Thereafter doping was performed with 0.2 nm of lithium and, finally, 150 nm of aluminium was vapour-deposited as the cathode and a 5 x 5 mm square device produced. The ionization potential of the electron transporting layer was 5.97 eV, the molecular weight was 609 and the glass transition temperature was 112°C. The ionization potential of the emissive layer was 5.79 eV, and so the difference in ionization potential in terms of the emissive layer was 0.18 eV.



ETM2

Comparative Example 4

**[0074]** A light emitting device was constructed in exactly the same way as in Example 24 except that the thickness of the Alq3 emissive layer was made 50 nm and no electron transporting material was used.

Comparative Example 5

**[0075]** A light emitting device was constructed in exactly the same way as in Example 24 except that BCP was used as the electron transporting material.

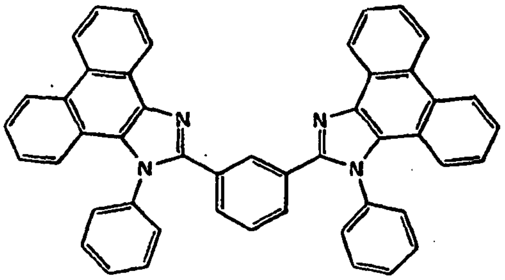
Example 25

**[0076]** A light emitting device was constructed in exactly the same way as in Example 24 except that the BQ-1 of Example 4 was used as the electron transporting material. The ionization potential of the electron transporting layer was 6.09 eV, the molecular weight was 670 and the glass transition temperature was 165°C. The difference in ionization potential in terms of the emissive layer was 0.30 eV.

**[0077]** The results obtained in Examples 24 and 25 are shown in Table 1 together with those of the comparative examples. It can be seen that the benzoquinoline derivative of the present invention also functioned effectively as the electron transporting material.

Example 26

**[0078]** A light emitting device was constructed in exactly the same way as in Example 24 except that there was used EM4, which is shown below, as the emissive material. At a driving voltage of 15 V, from this light emitting device there was obtained a blue coloured light emission based on the EM4 of peak emission wavelength 465 nm. The luminance was 1,200 cd/m$^2$ and the luminance efficiency was 1.0 cd/A. Thus, the benzoquinoline derivative of the present invention also functioned effectively as an electron transporting material for a blue coloured light emitting device.



EM4

Comparative Example 6

**[0079]** A light emitting device was constructed in exactly the same way as in Example 26 except that the thickness of the EM4 emissive material was made 50 nm and no electron transporting material was employed. At a driving voltage of 15 V, from this light emitting device there was obtained blue coloured light emission based on the EM4 of peak emission wavelength 465 nm. The luminance was 110 cd/m$^2$ and the luminance efficiency was 0.1 cd/A.

Example 27

**[0080]** A light emitting device was constructed in exactly the same way as in Example 24 except that, as the emissive layer region, co-deposition was carried out using tris(5,7-diphenyl-8-quinolinolato)aluminium(III) (A1(dPhq)3) as the host material and 4,4-difluoro-1,3,5,7-tetraphenyl-4-bora-3a,4a-diaza-indacene (PM-2) as the dopant material to a thickness of 15 nm, such that there was 1.0 wt% of the dopant. At a driving voltage of 10 V, from this light emitting device there was obtained red coloured light emission based on the dopant material of peak emission wavelength 615 nm. Thus, the benzoquinoline of the present invention functioned effectively as an electron transporting material for a red light emitting device.

Comparative Example 7

**[0081]** A light emitting device was constructed in exactly the same way as in Example 27 except that Alq3 was used as the electron transporting material. At a driving voltage of 10 V, red coloured light emission was not obtained from this light emitting device and an orange coloured light emission was produced, of peak emission wavelength 615 nm and having a shoulder peak in the region of 535 nm.

Example 28

**[0082]** A light emitting device was constructed in the same way as in Example 13 except that there was used the BQ-2 of Example 5 as the electron transporting layer. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the dopant material of peak emission wavelength 523 nm, and the luminance was 20,000 cd/$m^2$. The benzoquinoline derivative of the present invention functioned effectively as an electron transporting material for a green light emitting device.

Example 29

**[0083]** A light emitting device was constructed in the same way as in Example 16 except that there was used the BQ-3 of Example 5 as the electron transporting layer. At a driving voltage of 14 V, from this light emitting device there was obtained red coloured light emission based on the dopant material of peak emission wavelength 618 nm, and the luminance was 7,500 cd/$m^2$. The benzoquinoline derivative of the present invention functioned effectively as an electron transporting material for a red light emitting device.

Example 30

**[0084]** A light emitting device was constructed in the same way as in Example 13 except that for the emissive layer region there was used tris(8-quinolinolato)aluminium (III) (Alq3) as the host material and 3-(2-benzothiazolyl)-7-diethylaminocoumarin (Coumarin 2) as the dopant material, and a layer of ETM2 was provided of thickness 25 nm as the electron transporting layer. The ionization potential of the emissive layer was 5.78 eV and the ionization potential of the electron transporting layer was 5.97 eV, so the difference in ionization potentials between the electron transporting layer and the emissive layer was 0.19 eV. Furthermore, the molecular weight of the ETM2 was 609 and the glass transition temperature was 112°C. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the dopant material of peak emission wavelength 513 nm, and the luminance was 5,000 cd/$m^2$. Moreover, the percentage retention of the initial luminance after the elapse of 500 hours operation of this light emitting device was 70%, and a uniform emissive surface was maintained.

Comparative Example 8

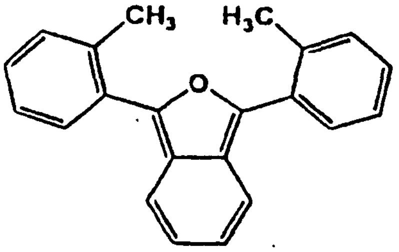
**[0085]** A light emitting device was constructed in exactly the same way as in Example 30 except that there was used Alq3 as the electron transporting layer. The ionization potential of this electron transporting layer was 5.79 eV and the difference in the ionization potentials of the electron transporting layer and emissive layer was 0.01 eV. Furthermore, the molecular weight of the Alq3 is 459 and its glass transition temperature 180°C. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the dopant material of peak emission wavelength 513 nm, and the luminance was 3,000 cd/$m^2$.

Comparative Example 9

**[0086]** A light emitting device was constructed in exactly the same way as in Example 30 except that there was used 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) as the electron transporting layer. The ionization potential of this electron transporting layer was 6.20 eV and the difference in the ionization potentials of the electron transporting layer and emissive layer was 0.42 eV. Furthermore, the molecular weight of BCP is 360 and its crystallization temperature 77°C. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the dopant material of peak emission wavelength 523 nm, and the luminance was 8,000 cd/$m^2$. However, the percentage retention of the initial luminance after the elapse of 500 hours operation of this light emitting device was below 50%, and unevenness of the emissive surface was observed.

Example 31

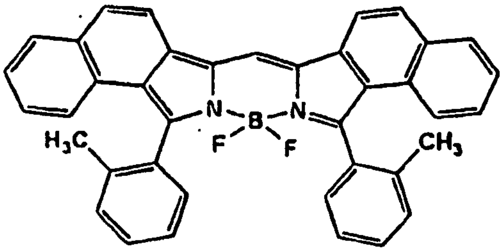
**[0087]** A light emitting device was constructed in exactly the same way as in Example 30 except that for the emissive layer region there was used EM4 as the host material and EM5, which is shown below, as the dopant material. The ionization potential of this emissive layer was 5.65 eV and the difference in the ionization potentials of the electron transporting layer and the emissive layer was 0.32 eV. At a driving voltage of 15 V, from this light emitting device there was obtained blue coloured light emission based on the dopant material of peak emission wavelength 477 nm, and the luminance was 3,500 cd/$m^2$. Thus, the electron transporting material of the present invention also functioned effectively in a blue light emitting device.



EM5

Example 32

**[0088]** A light emitting device was constructed in exactly the same way as in Example 30 except that for the emissive layer region there was codeposited, to a thickness of 15 nm, 1,4-diketo-2,5-bis(3,5-di-tert-butylbenzyl)-3,6-bis(4-biphenyl) pyrrolo[3,4-c]pyrrole (DPP-7) as the host material and EM6, which is shown below, as the dopant material, such that there was 1.0 wet% of the dopant. The ionization potential of the emissive layer was 5.79 eV and the difference in the ionization potentials of the electron transporting layer and the emissive layer was 0.18 eV. At a driving voltage of 14 V, from this light emitting device there was obtained red coloured light emission based on the dopant material of peak emission wavelength 629 nm, and the luminance was 8,000 cd/m$^2$. Thus, the electron transporting material of the present invention also functioned effectively in a red light emitting device.



EM6

Example 33

**[0089]** A light emitting device was constructed in exactly the same way as in Example 32 except that there was used BQ-1 of Example 4 as the electron transporting layer. The ionization potential of the BQ-1 was 6.09 eV and the difference in the ionization potentials of the electron transporting layer and the emissive layer was 0.30 eV. Furthermore, the molecular weight of BQ-1 was 670, the glass transition temperature was 165°C and no cold crystallization temperature was observed. At a driving voltage of 14 V, from this light emitting device there was obtained red coloured light emission based on the dopant material of peak emission wavelength 629 nm, and the luminance was 9,000 cd/m$^2$. Thus, the electron transporting material of the present invention also functioned effectively in a red light emitting device.

Comparative Example 10

**[0090]** A light emitting device was constructed in exactly the same way as in Example 32 except that there was used Alq3 as the electron transporting layer. The difference in the ionization potentials of the emissive layer and the electron transporting layer was 0 eV. At a driving voltage of 10 V, no red coloured light emission was obtained from this light emitting device. There was produced orange coloured light emission where there was a shoulder peak in the vicinity of 535 nm along with the peak emission wavelength of 629 nm.

Example 34

**[0091]** A light emitting device was constructed in exactly the same way as in Example 13 except that there was provided a layer of Alq3 of thickness 15 nm as the emissive layer and then there was provided a layer of the Phen-6 of Example 11 of thickness 35 nm as the electron transporting layer. At a driving voltage of 10 V, from this light emitting device there was obtained green coloured light emission based on the Alq3 of peak emission wavelength 536 nm, and the luminance was 4,000 cd/m$^2$. Furthermore, the percentage retention of the initial luminance after the elapse of 500 hours operation of this light emitting device was 75%, and a uniform emissive surface was maintained.

Example 35

**[0092]** Stages prior to the vapour-deposition were carried out in the same way as in Example 13. Then, by means of a resistance heating method, 20 nm of CuPc was vapour-deposited as the hole injecting material and 100 nm of $\alpha$-NPD as the hole transporting material. Next, as the emissive material a layer of Alq3 was provided of thickness of 50 nm. Then, a layer of the Phen-1 of Example 6 was provided of thickness 100 nm as the electron transporting material. Thereafter, there was 0.5 nm lithium doping onto the organic layer, following which 200 nm of aluminium was evaporated as the cathode, and a 5 x 5 mm square device produced. Film thicknesses were the values displayed by means of a quartz crystal oscillator type film thickness monitor. The ionization potential of the electron transporting layer was 6.09 eV, its molecular weight was 689 and the glass transition temperature was 197°C. The ionization potential of the emissive layer was 5.79 eV, and so the difference in ionization potentials was 0.30 eV. From this light emitting device, there was obtained high luminance green coloured light emission of emission wavelength 530 nm and luminance 20,000 cd/m$^2$. This light emitting device retained at least 80% of its luminance over a 1000 hour period. Furthermore, when this light emitting device was subjected to 1 mA pulse driving (duty ratio 1/60, 60 mA DC at time of pulse) within a vacuum cell, excellent light emission was confirmed.

Example 36

**[0093]** A light emitting device was constructed in exactly the same way as in Example 35 except that, for the emissive material, Alq3 was employed as the host material and 4-(dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) as the guest material, such that the dope concentration was 2%. From this light emitting device, there was obtained high luminance red-orange coloured light emission of emission wavelength 630 nm and luminance 10,000 cd/m$^2$. This light emitting device retained at least 80% of its luminance over a 1000 hour period.

Example 37

**[0094]** A light emitting device was constructed in exactly the same way as in Example 35 except that, as the host material, there was used 4,4'-bis(diphenylvinyl)biphenyl (DPVBi). From this light emitting device, there was obtained high luminance blue coloured light emission of emission wavelength 460 nm and luminance 10,000 cd/m$^2$.

Example 38

**[0095]** A light emitting device was constructed in exactly the same way as in Example 35 except that, as the host material, there was used DPP-1 and, as the guest material, there was used PM-2, such that the dope concentration was 1%. From this light emitting device, there was obtained a high luminance red coloured light emission of emission wavelength 610 nm and luminance 10,000 cd/m$^2$.

Comparative Example 11

**[0096]** A light emitting device was constructed in exactly the same way as in Example 38 except that there was used Alq3 as the electron transporting material. From this light emitting device, in addition to the red light emission from the emissive material there was observed green light emission from the electron transporting material, so the chromatic purity was markedly impaired.

Example 39

**[0097]** A light emitting device was constructed in exactly the same way as in Example 35 except that, as the host material, there was used 1,4-diketo-2,5-bis(3,5-di-tert-butylbenzyl)-3,6-bis(3-methoxyphenyl)pyrrolo[3,4-c]pyrrole (DPP-8) and, as the guest material, there was used 4,4-difluoro-1,3,5,7-tetra(4-methylphenyl)-8-phenyl-4-bora-3a,4a-diaza-indacene (PM-3), such that the dope concentration was 1%. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 625 nm and luminance 10,000 cd/m$^2$.

Example 40

**[0098]** A light emitting device was constructed in exactly the same way as in Example 35 except that, as the host material, there was used DPP-7 and, as the guest material, there was used 4,4-difluoro[3-phenyl-1-[(3-phenyl-2H-benzo[c]isoindol-1-yl)methylene-1H-benzo[c]isoindolato-N1,N2]borane (PM-4), such that the dope concentration was 1%. From this light emitting device, there was obtained a high luminance red coloured light emission of emission wavelength

635 nm and luminance 10,000 cd/m$^2$.

Example 41

**[0099]** A light emitting device was constructed in exactly the same way as in Example 35 except that, as the host material there was used DPP-2 and, as the guest material, there was used 4,4-difluoro-1,3,5,7-tetra(4-hexylphenyl)-4-bora-3a,4a-diaza-indacene (PM-5), such that the dope concentration was 1%. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 629 nm and luminance 10,000 cd/m$^2$.

Example 42

**[0100]** A light emitting device was constructed in exactly the same way as in Example 35 except that there was used DPP-3 as the host material and EM2 as the guest material, such that the dope concentration was 1%. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 615 nm and luminance 10,000 cd/m$^2$.

Example 43

**[0101]** A light emitting device was constructed in exactly the same way as in Example 35 except that there was used DPP-5 as the host material and EM3 as the guest material, such that the dope concentration was 1%. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 620 nm and luminance 10,000 cd/m$^2$.

Example 44

**[0102]** A light emitting device was constructed in exactly the same way as in Example 35 except that there was used 1,4-diketo-2,5-bis(3,5-dimethylbenzyl)-3,6-bis(4-ethylphenyl)pyrrolo[3,4-c]pyrrole (DPP-9) as the host material and 4,4-difluoro-1,3,5,7-tetra(4-butylphenyl)-8-phenyl-4-bora-3a,4a-diaza-indacene (PM-6) as the guest material, such that the dope concentration was 1%. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 615 nm and luminance 10,000 cd/m$^2$.

Example 45

**[0103]** A light emitting device was constructed in exactly the same way as in Example 35 except that there was used the Phen-2 of Example 7 as the electron transporting material. The ionization potential of the electron transporting layer was 6.11 eV, its molecular weight was 677 and the glass transition temperature was 175°C. The ionization potential of the emissive layer was 5.79 eV and so the difference in ionization potentials was 0.32 eV. From this light emitting device, there was obtained high luminance green coloured light emission of emission wavelength 530 nm and luminance 20,000 cd/m$^2$. This light emitting element maintained at least 80% of its luminance over a 1000 hour period. Furthermore, when this light emitting device was subjected to 1 mA pulse driving (duty ratio 1/60, 60 mA DC at time of pulse) within a vacuum cell, excellent light emission was confirmed.

Example 46

**[0104]** A light emitting device was constructed in exactly the same way as in Example 36 except that there was used the Phen-2 of Example 7 as the electron transporting material. From this light emitting device, there was obtained high luminance red-orange coloured light emission of emission wavelength 630 nm and luminance 10,000 cd/m$^2$. This light emitting element maintained at least 80% of its luminance over a 1000 hour period.

Example 47

**[0105]** A light emitting device was constructed in exactly the same way as in Example 37 except that there was used the Phen-2 of Example 7 as the electron transporting material. From this light emitting device, there was obtained high luminance blue coloured light emission of emission wavelength 460 nm and luminance 10,000 cd/m$^2$.

Example 48

**[0106]** A light emitting device was constructed in exactly the same way as in Example 38 except that there was used the Phen-2 of Example 7 as the electron transporting material. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 610 nm and luminance 10,000 cd/m$^2$.

Example 49

**[0107]** A light emitting device was constructed in exactly the same way as in Example 44 except that there was used the TPM-dNTR of Example 12 as the electron transporting material. From this light emitting device, there was obtained high luminance red coloured light emission of emission wavelength 615 nm and luminance 10,000 cd/m$^2$.

Example 50

**[0108]** A glass substrate on which a 150 nm ITO transparent electroconductive film had been deposited (produced by the Asahi Glass Co., 15 Ω/□, electron beam vapour-deposited product) was cut to 30 x 40 mm and, by a photolithography method, pattern processing was carried out in the form of 32 stripes x 300 μm pitch (remaining width 270 μm). In order to facilitate external electrical connection, the ITO stripes were widened to a 1.27 mm pitch (width of opening portion 800 μm) at one side in the lengthwise direction. The substrate obtained was subjected to ultrasonic washing for 15 minute periods with acetone and "Semico-Clean 56" respectively, after which washing was carried out with ultrapure water. Next, 15 minutes ultrasonic washing was performed with isopropyl alcohol, after which it was immersed for 15 minutes in hot methanol and dried. Just prior to producing the device, this substrate was given a 1 hour UV-ozone treatment, then placed in vacuum vapour-deposition apparatus and the apparatus evacuated until the degree of vacuum inside was less than 5 x 10$^{-4}$ Pa. By means of a resistance heating method, firstly there was vapour-deposited 10 nm of CuPc, and then there was vapour-deposited 50 nm of α-NPD. Next there was codeposited to a thickness of 25 nm the emissive layer region, using Alq3 as the host material and Coumarin-1 as the dopant material, such that there was 1.0 wt% of dopant. Thereafter, there was provided a ETM1 layer of thickness 25 nm as the electron transporting layer. The ionization potential of the electron transporting layer was 6.07 eV, the molecular weight was 672, the glass transition temperature 219°C and the cold crystallization temperature 352°C. Next, under vacuum, mask exchange was performed with a mask comprising a kovar sheet of thickness 50 μm in which sixteen 250 μm apertures (remaining width 50 μm, corresponding to a 300 μm pitch) had been provided by wet etching, so as to intersect the ITO stripes at right angles, and then this fixed with a magnet from the underside so that the mask and ITO substrate closely adhered. After doping the organic layer with 0.5 nm of lithium, there was vapour-deposited 200 nm of aluminium and a 32 x 16 dot matrix device produced. When this device was subjected to matrix driving, character display was possible without cross-talk.

Example 51

**[0109]** A 32 x 16 dot matrix device was constructed in exactly the same way as in Example 50 except that there was used the Phen-6 of Example 11 as the electron transporting layer. When this device was subjected to matrix driving, character display was possible without cross-talk.

Example 52

**[0110]** A 32 x 16 dot matrix device was constructed in exactly the same way as in Example 50 except that a 25 nm layer of Alq3 was provided as the emissive layer and a 25 nm layer of ETM2 was provided as the electron transporting layer. When this device was subjected to matrix driving, character display was possible without cross-talk.

Example 53

**[0111]** The stages up to the vapour-deposition were carried out in the same way as in Example 50. Then, by means of a resistance heating method, there was vapour-deposited 150 nm of 4,4'-bis(N-(m-tolyl)-N-phenylamino)biphenyl as the hole transporting material, and there was evaporated 50 nm of Alq3 as the emissive material. Next, as the electron transporting material there was provided the Phen-1 of Example 6 at a layer thickness of 100 nm. The film thicknesses were the values displayed by means of a quartz crystal oscillator type film thickness monitor. Then, under vacuum, mask exchange was performed with a mask comprising a kovar sheet of thickness 50 μm in which sixteen 250 μm apertures (remaining width 50 μm, corresponding to a 300 μm pitch) had been provided by wet etching, so as to intersect the ITO stripes at right angles, and this then fixed with a magnet from the underside so that the mask and ITO substrate closely adhered. After doping the organic layer with 0.5 nm of lithium, there was vapour-deposited 200 nm of aluminium and a 32 x 16 dot matrix device produced. When this device was subjected to matrix driving, character display was possible without cross-talk.

Example 54

**[0112]** A 32 x 16 dot matrix device was constructed in exactly the same way as in Example 53 except that there was used the Phen-2 of Example 7 as the electron transporting layer. When this device was subjected to matrix driving, character display was possible without cross-talk.

Industrial Application Potential

**[0113]** The present invention provides a light emitting device of outstanding thermal stability, high electrical energy utilization efficiency and excellent chromatic purity, together with light emitting device materials for use therein.

Table 1

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 1 | basic spiro structure synthesis (1) | 9 | | | | | | | | | | |
| Example 2 | basic spiro structure synthesis (2) | 9 | | | | | | | | | | |
| Example 3 | introduction of reactive substituents into basic spiro structure | 9 | | | | | | | | | | |
| Example 4 | benzoquinoline derivative synthesis (BQ-1) | (3) | | | | | | | | | | |
| Example 5 | benzoquinoline derivative syntheses (BQ-2,3) | (3) | | | | | | | | | | |
| Example 6 | phenanthroline derivative synthesis (Phen-1) | 9 | | | | | | | | | | |
| Example 7 | phenanthroline derivative synthesis (Phen-2) | 10 | | | | | | | | | | |

(continued)

|  | Structure | Corresponding Claim or Corresponding Example | Results |  |  |  |  |  |  |  |  |  |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
|  |  |  | Properties |  |  |  | EL Device |  |  |  |  |  |
|  |  |  | Electron Transporting Layer |  |  | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
|  |  |  | Mw | Tg (°C) | IP [eV] | [eV] |  | [nm] | [cd/m$^2$] | [cd/A] | [%] |  |
| Example 8 | phenanthroline derivative synthesis (Phen-3) | (5) |  |  |  |  |  |  |  |  |  |  |
| Example 9 | phenanthroline derivative synthesis (Phen-4) | (5) |  |  |  |  |  |  |  |  |  |  |
| Example 10 | phenanthroline derivative synthesis (Phen-5) | (5) |  |  |  |  |  |  |  |  |  |  |
| Example 11 | 1,7-phenanthroline derivative synthesis (Phen-6) | (5) |  |  |  |  |  |  |  |  |  |  |
| Example 12 | naphthyridine derivative synthesis (TPM-dNTR) | 10 |  |  |  |  |  |  |  |  |  |  |
| Example 13 | Alq3+Coumarin-1 (green)/ETM1 (phenanthroline multimer) | 4-8 | 672 | 219 | 6.07 | 0.29 | green | 523 | 38,000(10V) |  | 80 | uniform |
| Comp.Ex.1 | Alq3+Coumarin-1 (green)/Alq | Example 13 | 459 | 180 | 5.79 | 0.01 | green | 523 | 6,000(10V) |  |  |  |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Comp.Ex.2 | Alq3+Coumarin-1 (green)/BCP | Example 13 | 360 | <77 | 6.2 | 0.42 | green | 523 | 12,000(10V) | | <50 | uneven |
| Example 14 | carbazole+ phosphorescent guest (green) /ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | green | 515 | 1.000(6V) | | | |
| Example 15 | EM1(blue)/ETM1 (phenanthroline multimer) | 4,5,7 | 672 | 219 | 6.07 | | blue | 463 | 8,000(15V) | | | |
| Example 16 | DPP-1+EM2(red)/ ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 618 | 10,000 | | | |
| Comp.Ex.3 | DPP-1+EM2 (red) /Alq | Examples 16-22 | 459 | 180 | 5.79 | | orange | 618,535 | | | | |
| Example 17 | DPP-1+EM2(red)/ Phen-3 (phenanthroline multimer) | 4,5,7,8 | 663 | 150 | 6.14 | | red | 618 | 7,000 | | | |
| Example 18 | DPP-2+EM2(red)/ ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 618 | 8,500 | | | |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 19 | DPP-3+EM2 (red)/ ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 618 | 6,500 | | | |
| Example 20 | DPP-4+EM2 (red)/ ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 618 | 7,000 | | | |
| Example 21 | DPP-5+EM3 (red)/ ETM1 (phenanthioline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 619 | 8,900 | | | |
| Example 22 | DPP-6+EM3 (red)/ ETM1 (phenanthroline multimer) | 4,5,7,8 | 672 | 219 | 6.07 | | red | 619 | 6,600 | | | |
| Example 23 | BQ-1 (benzoquinoline multimer)+PM-1 (green)/BQ-1 | 1(b),2,3 | | | | | green | 519 | 8,000(IIV) | | | |
| Example 24 | Alq3 (green) /ETM2 (benzoquinoline multimer) | 1(b),2-4,6,7 | 609 | 112 | 5.97 | 0.18 | green | 536 | 4,000 | 2.0 | 80 | uniform |
| Comp.Ex.4 | Alq3 (green)/Alq | Examples 24, 25 | 459 | 180 | 5.79 | 0 | green | 536 | 3,800 | 1.9 | | |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Comp.Ex.5 | Alq3 (green)/BCP | Examples 24, 25 | 360 | <77 | 6.2 | 0.41 | green | 536 | 3,000 | 1.4 | <50 | uneven |
| Example 25 | Alq3 (green)/BQ-1 (benzoquinoline multimer | 1(b),2-4,6,7 | 670 | 165 | 6.09 | 0.30 | green | 536 | 3,500 | 1.8 | 80 | uniform |
| Example 26 | EM4 (blue)/ETM2 (benzoquinoline multimer | 1(b),2-4,7 | 609 | 112 | 5.97 | | blue | 465 | 1200(15V) | 1.0 | | |
| Comp.Ex.6 | EM4 (blue) | Example 26 | | | | | blue | 465 | 110(15V) | 0.1 | | |
| Example 27 | Al(dPhq)3+PM-2 (red)/ ETM2 (benzoquinoline multimer | 1(b),2-4,7-8 | 609 | 112 | 5.97 | | red | 615 | | | | |

Note 1) The structures in Examples 13 to 27 relate to just the emissive layer/electron transporting layer portion of the device structure

Note 2) Mw = molecular weight, Tg = glass transition temperature, IP = ionization potential, IP difference = difference in ionization potentials between the electron transporting layer and emissive layer

Table 2

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/$m^2$] | [cd/A] | [%] | |
| Comp.Ex.7 | Al(dPhq)3+PM-2 (red) /Alq | Example 27 | 459 | 180 | 5.79 | | orange | 615,535 | | | | |
| Example 28 | Alq3+Coumarin-1 (green)/BQ-2 (benzoquinoline multimer) | 1(b),2,3 | | | | | green | 523 | 20,000(10V) | | | |
| Example 29 | DPP-1+EM2 (red)/BQ-3 (benzoquinoline multimer) | 1(b),2,3 | | | | | red | 618 | 7,500(14V) | | | |
| Example 30 | Alq3+Coumarin-2 (green)/ETM2 (benzoquinoline multimer) | 1(b),2-4,6-8 | 609 | 112 | 5.97 | 0.19 | green | 513 | 5,000(10V) | | 70 | uniform |
| Comp.Ex.8 | Alq3+Coumarin-2 (green)/Alg | Example 30 | 459 | 180 | 5.79 | 0.01 | green | 513 | 3,000(10V) | | | |
| Comp.Ex.9 | Alq3+Coumarin-2 (green)/BCP | Example 30 | 360 | <77 | 6.20 | 0.42 | green | 513 | 8.000(10V) | | <50 | uneven |
| Example 31 | EM4+EM5 (blue)/ ETM2 (benzoquinoline multimer) | 1(b),2-4,6-8 | 609 | 112 | 5.97 | 0.32 | blue | 477 | 3,500(15V) | | | |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 32 | DPP-7+EM6 (red)/ ETM2 (benzoquinoline multimer) | 1(b),2-4,6-8 | 609 | 112 | 5.97 | 0.18 | red | 629 | 8,000(14V) | | | |
| Example 33 | DPP-7+EM6 (red)/ BQ-1 (benzoquinoline multimer) | 1(b),2-4,6-8 | 670 | 165 | 6.09 | 0.30 | red | 629 | 9,000(14V) | | | |
| Comp.Ex. 10 | DPP-7+EM6 (red)/ Alq | Examples 32, 33 | 459 | 180 | 5.79 | 0 | orange | 629,535 | | | | - |
| Example 34 | Alq (green)/Phen-6 (1,7-phenanthroline multimer) | 1(a) | | | | | green | 536 | 4,000(10V) | | 75 | uniform |
| Example 35 | Alq (green)/Phen-1 (spiro phenanthroline multimer) | 1(c),4-7 | 689 | 197 | 6.09 | 0.30 | green | 530 | 20,000 | | >80 | |
| Example 36 | Alq+DCJTB (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 630 | 10,000 | | >80 | |
| Example 37 | DPVBi (blue)/Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7 | 689 | 197 | 6.09 | | blue | 460 | 10,000 | | | |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 38 | DPP-1+PM-2 (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 610 | 10,000 | | | |
| Comp.EX. 11 | DPP-1+PM-2 (red)/Alq | Example 38 | 459 | 180 | 5.79 | | red + green | | | | | |
| Example 39 | DPP-8+PM-3 (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 625 | 10,000 | | | |
| Example 40 | DPP-7+PM-4 (red)/ Phen-1 (spiro phenanthroline multimer) | 1 (c),4,5,7,8 | 689 | 197 | 6.09 | | red | 635 | 10,000 | | | |
| Example 41 | DPP-2+PM-5 (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 629 | 10,000 | | | |
| Example 42 | DPP-3+EM2 (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 615 | 10,000 | | | |
| Example 43 | DPP-5+EM3 (red)/ Phen-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 620 | 10,000 | | | |

(continued)

| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 44 | DPP-9+PM-6 (red)/ Pben-1 (spiro phenanthroline multimer) | 1(c),4,5,7,8 | 689 | 197 | 6.09 | | red | 615 | 10,000 | | | |
| Example 45 | Alq (green)/Phen-2 (tetraphenylmethane phenanthroline multimer) | 1(d),4-7 | 677 | 175 | 6.11 | 0.32 | green | 530 | 20,000 | | | |
| Example 46 | Alq+DCJTB (red)/ Phen-2 (tetraphenylmethane phenanthroline multimer) | 1(d),4,5,7,8 | 677 | 175 | 6.11 | | red | 630 | 10,000 | | | |
| Example 47 | DPVBi (blue)/Phen-2 (tetraphenyl-methane phenandiroline multimer) | 1(d),4,5,7 | 677 | 175 | 6.11 | | blue | 460 | 10,000 | | | |
| Example 48 | DPP-1+PM-2 (red)/ Phen-2 (tetraphenylmethane phenanthroline multimer) | 1(d),4,5,7,8 | 677 | 175 | 6.11 | | red | 610 | 10,000 | | | |
| Example 49 | DPP-9+PM-6 (red) /TPM-dNTR (tetraphenylmethane derivative) | 1(d),4,5,7,8 | 677 | 175 | 6.11 | | red | 615 | 10,000 | | | |

(continued)

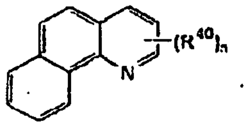
| | Structure | Corresponding Claim or Corresponding Example | Results | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Properties | | | | EL Device | | | | | |
| | | | Electron Transporting Layer | | | IP difference | Colour | Wavelength | Luminance (voltage) | Efficiency | Retention | Emissive Surface |
| | | | Mw | Tg (°C) | IP [eV] | [eV] | | [nm] | [cd/m$^2$] | [cd/A] | [%] | |
| Example 50 | dot matrix (using ETM1 (phenanthroline multimer)) | 11 | | | | | | | | | | |
| Example 51 | dot matrix (using Phen-6 (1,7-phenanthroline multimer)) | 11 | | | | | | | | | | |
| Example 52 | dot matrix (using ETM2 (benzoquinoline multimer)) | 11 | | | | | | | | | | |
| Example 53 | dot matrix (using Phen-1 (spiro-phenanthroline multimer)) | 11 | | | | | | | | | | |
| Example 54 | dot matrix (using Phen-2 (tetraphenylmethane-phenanthroline multimer)) | 11 | | | | | | | | | | |

Note 1) The structures in Examples 28 to 49 relate to just the emissive layer/electron transporting layer portion of the device structure
Note 2) Examples 50 to 54 are examples of dot matrixes, with only the compound of the present invention used being noted
Note 3) Mw = molecular weight, Tg = glass transition temperature, IP = ionization potential, IP difference = difference in ionization potentials of the electron transporting layer and emissive layer

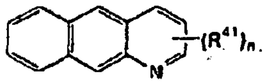
## Claims

1. A light emitting device which is **characterized in that** it is a device having a structure in which at least an anode, an emissive layer, an electron transporting layer and a cathode are provided as layers in turn, and which emits light by means of electrical energy, and said device has a benzoquinoline derivative, where said benzoquinoline derivative has a plurality of benzoquinoline skeletal structure, and the plurality of benzoquinoline skeletal structures are connected by a connecting unit which contains a conjugated bond or conjugated bonds, an aromatic hydrocarbon, an aromatic heterocycle or mixture thereof.

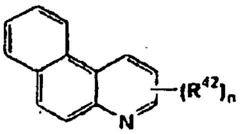
2. The light emitting device of claim 1, wherein each benzoquinoline skeletal structure is independently represented by one of general formulae (9) to (14):



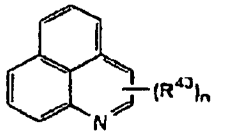
(9)



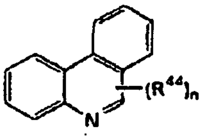
(10)



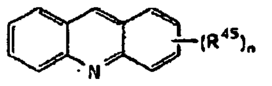
(11)



(12)



(13)



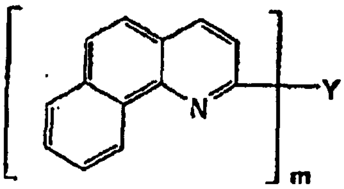
(14)

$R^{40}$ to $R^{45}$ in general formulae (9) to (14) denote substituent groups at any position in the benzoquinoline skeletal structure other than a position used for connection to another benzoquinoline skeletal structure, and are selected from amongst hydrogen,

alkyl groups, cycloakyl groups, aralkyl groups, alkenyl groups, cycloalkenyl groups, alkoxy groups, alkylthio groups where the oxygen atom of the ether linkage in an alkoxy group is replaced by a sulphur atom, aryl ether groups, aryl thioether groups where the oxygen atom of the ether linkage in an aryl ether group is replaced by a sulphur atom, aryl groups, heterocyclic groups, halogens, cyano group, aldehyde group, carbonyl group, carboxyl group, ester group, carbamoyl group, amino group, nitro group, silyl groups, siloxanyl groups which are groups with silicon having an interposed ether linkage, and cyclic structures formed with an adjacent substituent group, n is an integer in the range 1 to 8 and, in the case where there are a plurality of substituents, said substituents may be the same or different.

3. The light emitting device according to claim 2 wherein the benzoquinoline derivative is represented by general

formula (15).



(15)

where Y is a connecting unit which contains a conjugated bond or conjugated bonds, an aromatic hydrocarbon, an aromatic heterocycle or mixture thereof, and m is 2 or more.

4. A light emitting device according to any one of claims 1 to 3, where the organic compound from which the electron transporting layer is composed is an organic compound of molecular weight at least 400, glass transition temperature at least 90°C, ionization potential at least 5.9 eV and having a benzoquinoline skeletal structure.

5. A light emitting device according to any one of claims 1 to 4 which is **characterized in that** the electron transporting layer has an ionization potential which is at least 0.1 eV higher than the ionization potential of the emissive layer.

6. A light emitting device according to any one of claims 1 to 4 where the materials from which the emissive layer and the electron transporting layer are composed are organic compounds which are sublimable.

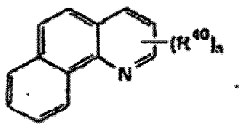
7. A light emitting device according to any one of claims 1 to 4 where the emissive layer is composed of at least two types of organic compound.

8. A display in which a light emitting device according to any one of claims 1 to 4 is made to display by a matrix and/or segment system.

## Patentansprüche

1. Lichtemittierende Vorrichtung, die **dadurch gekennzeichnet ist, dass** es sich um eine Vorrichtung mit einer Struktur handelt, in der zumindest eine Anode, eine Emissionsschicht, eine Elektronentransportschicht und eine Kathode als aufeinander folgende Schichten bereitgestellt sind, und die Licht mittels elektrischer Energie emittiert, und wobei die Vorrichtung ein Benzochinolinderivat aufweist, wobei das Benzochinolinderivat eine Vielzahl von Benzochinolin-Skelettstrukturen aufweist und die Vielzahl von Benzochinolin-Skelettstrukturen über eine Verbindungseinheit verbunden sind, die eine oder mehrere konjugierte Bindungen, einen aromatischen Kohlenwasserstoff, einen aromatischen Heterozyklus oder ein Gemisch davon umfasst.

2. Lichtemittierende Vorrichtung nach Anspruch 1, wobei die einzelnen Benzochinolin-Skelettstrukturen unabhängig voneinander durch eine der allgemeinen Formeln (9) bis (14) dargestellt sind:



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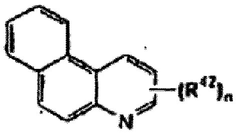

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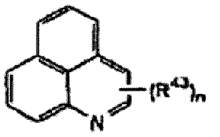

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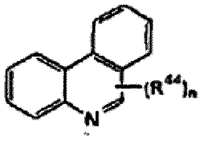

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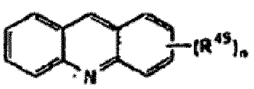
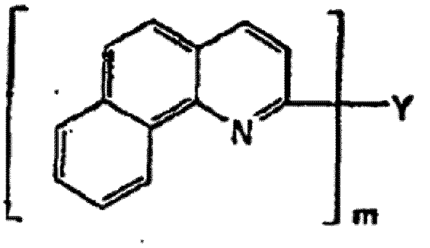


(14)

worin $R^{40}$ bis $R^{45}$ in den allgemeinen Formeln (9) bis (14) Substituentengruppen an beliebigen Positionen in der Benzochinolin-Skelettstruktur, die nicht als Position zur Verbindung mit einer anderen Benzochinolin-Skelettstruktur verwendet werden, bezeichnen und aus Wasserstoff, Alkylgruppen, Cycloalkylgruppen, Aralkylgruppen, Alkenylgruppen, Cycloalkenylgruppen, Alkoxygruppen, Alkylthiogruppen, in denen das Sauerstoffatom der Etherbindung in einer Alkoxygruppe durch ein Schwefelatom ersetzt ist, Arylethergruppen, Arylthioethergruppen, in denen das Sauerstoffatom der Etherbindung in einer Arylethergruppe durch ein Schwefelatom ersetzt ist, Arylgruppen, heterozyklischen Gruppen, Halogenen, Cyanogruppen, Aldehydgruppen, Carbonylgruppen, Carboxylgruppen, Estergruppen, Carbamoylgruppen, Aminogruppen, Nitrogruppen, Silylgruppen, Siloxanylgruppen, die Gruppen mit Silicium mit einer dazwischenliegenden Etherbindung sind, und mit einer benachbarten Substituentengruppe gebildeten zyklischen Strukturen ausgewählt sind, n eine ganze Zahl im Bereich von 1 bis 8 ist und, wenn es eine Vielzahl von Substituenten gibt, die Substituenten gleich oder unterschiedlich sein können.

3. Lichtemittierende Vorrichtung nach Anspruch 2, wobei das Benzochinolinderivat durch die allgemeine Formel (15) dargestellt ist:



(15)

worin Y eine Verbindungseinheit ist, die eine oder mehrere konjugierte Bindungen, einen aromatischen Kohlenwasserstoff, einen aromatischen Heterozyklus oder ein Gemisch davon enthält, und m = 2 oder mehr ist.

4. Lichtemittierende Vorrichtung nach einem der Ansprüche 1 bis 3, wobei die organische Verbindung, aus der die Elektronentransportschicht besteht, eine organische Verbindung mit einem Molekulargewicht von zumindest 400, einer Glastemperatur von zumindest 90 °C und einem Ionisierungspotential von zumindest 5,9 eV ist und eine Benzochinolin-Skelettstruktur aufweist.

5. Lichtemittierende Vorrichtung nach einem der Ansprüche 1 bis 4, die **dadurch gekennzeichnet ist, dass** die Elektronentransportschicht ein Ionsierungspotential aufweist, das zumindest 0,1 eV höher ist als das Ionisierungs-

potential der Emissionsschicht.

6. Lichtemittierende Vorrichtung nach einem der Ansprüche 1 bis 4, wobei die Materialien, aus denen die Emissionsschicht und die Elektronentransportschicht bestehen, organische Verbindungen sind, die sublimierbar sind.

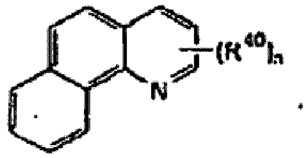
7. Lichtemittierende Vorrichtung nach einem der Ansprüche 1 bis 4, wobei die Emissionsschicht aus zumindest zwei Arten von organischen Verbindungen besteht.

8. Anzeige, in der eine lichtemittierende Vorrichtung nach einem der Ansprüche 1 bis 4 ausgebildet ist, um mittels einer Matrix und/oder ein Segmentsystem anzuzeigen.

## Revendications

1. Dispositif luminescent **caractérisé en ce qu'**il s'agit d'un dispositif ayant une structure dans laquelle au moins une anode, une couche émissive, une couche de transport d'électrons et une cathode sont disposées en couches tour à tour, et qui émet une lumière au moyen d'énergie électrique, ledit dispositif ayant un dérivé de benzoquinoline, ledit dérivé de benzoquinoline ayant une pluralité de structures squelettiques de benzoquinoline, et la pluralité de structures squelettiques de benzoquinoline étant connectées par un motif de connexion qui contient une liaison conjuguée ou des liaisons conjuguées, un hydrocarbure aromatique, un hétérocycle aromatique ou un de leurs mélanges.

2. Dispositif luminescent selon la revendication 1, dans lequel chaque structure squelettique de benzoquinoline est indépendamment représentée par l'une des formules (9) à (14) suivantes :



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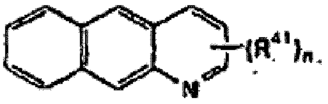

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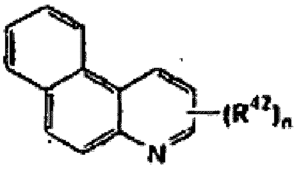

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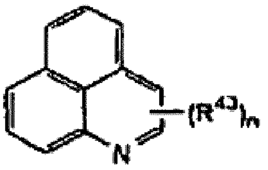

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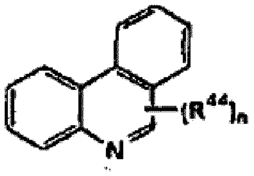

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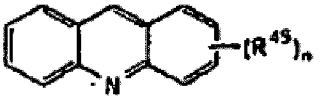
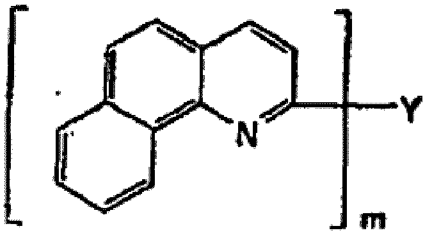


(14)

dans les formules générales (9) à (14), $R^{40}$ à $R^{45}$ désignent des groupes substituants en n'importe quelle position dans la structure squelettique de benzoquinoline autre qu'une position utilisée pour la connexion à une autre structure squelettique de benzoquinoline, et sont choisis parmi l'hydrogène, les groupes alkyle, les groupes cycloalkyle, les groupes aralkyle, les groupes alcényle, les groupes cycloalcényle, les groupes alcoxy, les groupes alkylthio où l'atome d'oxygène de la liaison éther dans un groupe alcoxy est remplacé par un atome de soufre, les groupes aryléther, les groupes arylthioéther où l'atome d'oxygène de la liaison éther dans un groupe aryléther est remplacé par un atome de soufre, les groupes aryle, les groupes hétérocycliques, les halogènes, un groupe cyano, un groupe aldéhyde, un groupe carbonyle, un groupe carboxyle, un groupe ester, un groupe carbamoyle, un groupe amino, un groupe nitro, les groupes silyle, les groupes siloxanyle qui sont des groupes avec du silicium ayant une liaison éther interposée, et les structures cycliques formées avec un groupe substituant adjacent, n est un entier situé dans la plage allant de 1 à 8, et, dans le cas où il y a plusieurs substituants, lesdits substituants peuvent être identiques ou différents.

3. Dispositif luminescent selon la revendication 2, dans lequel le dérivé de benzoquinoline est représenté par la formule générale (15) :



(15)

dans laquelle Y est un motif de connexion qui contient une liaison conjuguée ou des liaisons conjuguées, un hydrocarbure aromatique, un hétérocycle aromatique ou un de leurs mélanges, et m vaut 2 ou plus.

4. Dispositif luminescent selon l'une quelconque des revendications 1 à 3, dans lequel le composé organique composant la couche de transport d'électrons est un composé organique ayant une masse moléculaire d'au moins 400, une température de transition vitreuse d'au moins 90°C, un potentiel d'ionisation d'au moins 5,9 eV et ayant une structure squelettique de benzoquinoline.

5. Dispositif luminescent selon l'une quelconque des revendications 1 à 4, qui est **caractérisé en ce que** la couche de transport d'électrons a un potentiel d'ionisation qui est supérieur d'au moins 0,1 eV au potentiel d'ionisation de la couche émissive.

6. Dispositif luminescent selon l'une quelconque des revendications 1 à 4, dans lequel les matériaux composant la couche émissive et la couche de transport d'électrons sont des composés organiques qui sont sublimables.

7. Dispositif luminescent selon l'une quelconque des revendications 1 à 4, dans lequel la couche émissive est composée d'au moins deux types de composés organiques.

8. Affichage dans lequel un dispositif luminescent selon l'une quelconque des revendications 1 à 4 est conçu pour afficher au moyen d'un système à matrice et/ou à segments.

## REFERENCES CITED IN THE DESCRIPTION

### Patent documents cited in the description

- US 5393614 A **[0006]**
- US 5840217 A **[0034]**

### Non-patent literature cited in the description

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- *J. Am. Chem. Soc.,* 1930, vol. 52, 2881 **[0034]**
- *J. Am. Chem. Soc.,* 1958, vol. 80, 1883 **[0035]**
- *Libigs Ann. Chem.,* 1971, vol. 749, 38 **[0036]**
- *J. Org. Chem.,* 1996, vol. 61, 3021 **[0038]**

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|---|---|---|---|
| 专利名称(译) | 发光元件材料和包含其的发光元件 | | |
| 公开(公告)号 | EP2365735B1 | 公开(公告)日 | 2013-08-28 |
| 申请号 | EP2011169303 | 申请日 | 2001-11-22 |
| [标]申请(专利权)人(译) | 东丽株式会社 | | |
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| IPC分类号 | H05B33/22 H05B33/14 C09K11/06 C07D311/96 C07D471/04 C07D405/14 C07D409/14 C07D413/14 C07D417/14 C07D407/14 C07D335/12 C07C15/28 C07C15/52 C07D209/14 C07D209/86 C07D213/53 C07D215/12 C07D219/02 C07D221/10 C07D235/20 C07D241/42 C07D249/08 C07D263/32 C07D263/62 C07D263/64 C07D271/10 C07D271/107 C07D277/66 C07D519/00 C07F7/08 C07F9/38 C07F9/6561 H01L51/00 H01L51/30 H01L51/50 | | |
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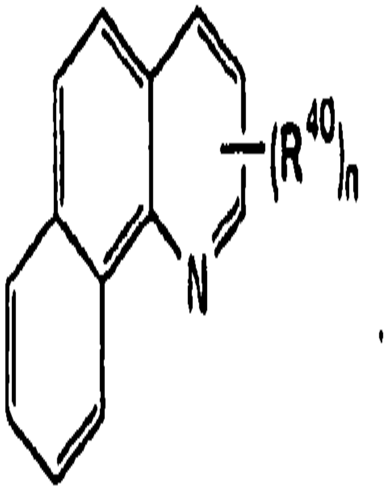
| 优先权 | 2000357129 2000-11-24 JP<br>2001173610 2001-06-08 JP | | |
| 其他公开文献 | EP2365735A3<br>EP2365735A2 | | |
| 外部链接 | Espacenet | | |

**摘要(译)**

发光器件的特征在于它是具有这样的结构的器件，其中至少阳极，发光层，电子传输层和阴极依次作为层提供，并且通过电能发光，所述装置具有苯喹啉衍生物。苯并喹啉衍生物可具有多个苯醌骨架结构，包括通过共轭键连接的多个苯醌骨架结构，芳烃，芳香杂环或其混合物。示例性发光器件具有这样的结构，其中至少阳极，发光层，电子传输层和阴极依次作为层提供并且通过电能发光，并且有机化合物是有机化合物。分子量至少为400，玻璃化转变温度至少为90℃，电离电位至少为5.9eV，并具有苯并喹啉骨架结构。



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