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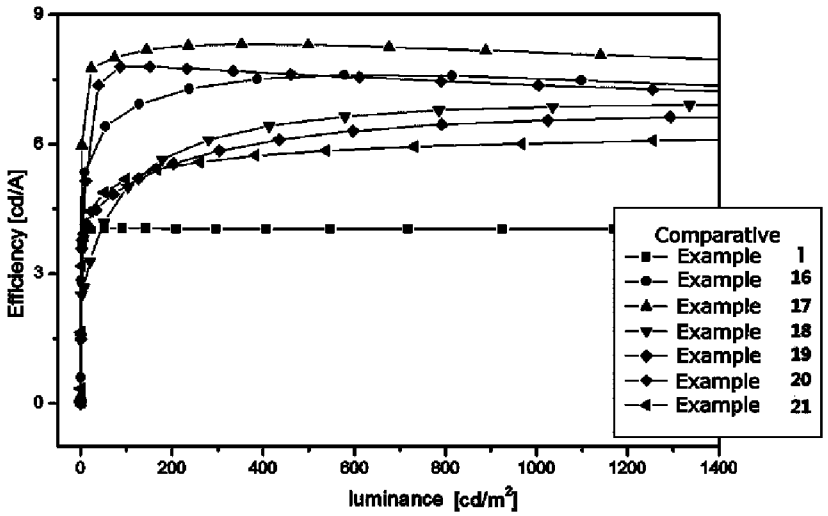
(54) **Title**: NEW ELECTRON TRANSPORT MATERIAL AND ORGANIC ELECTROLUMINESCENT DEVICE USING THE SAME



(57) **Abstract**: Provided are a new electron transport material and an organic electroluminescent device including the same. The electron transport material according to the present invention may have the excellent luminescence property and reduce the driving voltage to increase the power efficiency, such that the organic electroluminescent device using less consumption power may be manufactured.

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Description

Title of Invention: NEW ELECTRON TRANSPORT MATERIAL AND ORGANIC ELECTROLUMINESCENT DEVICE USING THE SAME

Technical Field

[1] The following disclosure relates to a new electron transport material and an organic electroluminescent device using the same.

Background Art

[2] An organic electroluminescent device is a device emitting light while electrons and holes disappear after being coupled in pairs when an electron charge is injected into an organic film formed between an electron injection electrode (cathode) and a hole injection electrode (anode). The organic electroluminescent device has advantages in that the device may be formed on a flexible transparent substrate made of, for example, plastic material, driven at a low voltage (10V or less) as compared with a plasma display panel or an inorganic electroluminescent (EL) display, has relatively low power consumption and excellent color sensation.

[3] Generally, the organic electroluminescent device has a structure consisting of a substrate, an anode, a hole injection layer receiving a hole from the anode, a hole transport layer transporting the hole, a luminescent layer emitting light while the hole and an electron are bonded, an electron transport layer receiving the electrode from a cathode to transport the electron to the luminescent layer, and the cathode. In some cases, the luminescent layer may be configured by applying a small amount of a fluorescent or phosphorescent dye to the electron transport layer or the hole transport layer without a separate luminescent layer. In the case of using a polymer, generally, one polymer may entirely serve as the hole transport layer, the luminescent layer, and the electron transport layer. Organic thin film layers between two electrodes may be formed by a method such as a vacuum deposition method, a spin coating method, an inkjet printing method, a roll coating method, or the like, and for effective injection of the electron from the cathode, a separate electron injection layer may be inserted.

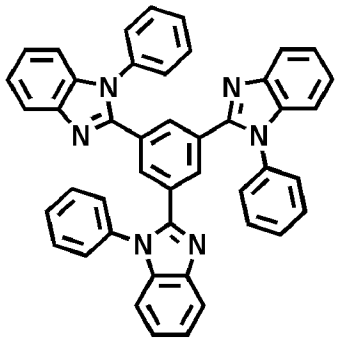
[4] In the case in which an interface between the electrode and an organic material is stabilized, or in the case of the organic material, since there is a large difference in a movement rate between the hole and the electron, an appropriate hole transport layer and electron transport layer are used, the hole and the electron may be effectively transported to the luminescent layer. In addition, in order to balance the hole and the electron densities in the luminescent layer to increase luminescence efficiency, the organic electroluminescent device is manufactured to have a multi-layer thin film

structure.

[5] Meanwhile, as a representative example of the existing electron transport material, there are aluminum complex and beryllium complex such as Alq3 (tris(8-hydroxyquinoline)aluminum(III)) and Bebq (bis(10-hydroxybenzo-[h]quinolinato)beryllium). However, in the case in which these materials are used in a blue electroluminescent device, color purity may be decreased due to luminescence caused by exciton diffusion.

[6] In addition, TPBI, which was reported by Kodak in 1996 disclosed in US Patent No. 5,645,948, (See the following structure), is known to be a representative material for electron transport layer having an imidazole group. This material contains three N-phenyl benzimidazole groups at 1, 3, and 5 substitution positions of benzene, and has a function of blocking holes from a luminescent layer as well as transporting electrons. However, stability of TPBI is too low to be actually used in the device.

[7]



TPBI

[8] In the case of the electron transport material according to the related art, unlike the reported contents, actually, the material may slightly improve only a driving voltage, or there are problems such as significant deterioration of a device driving lifespan or negative properties such as variation in the device lifespan according to the color, and deterioration of thermal stability, or the like.

[9] In addition, a fluorescent material was used in the organic electroluminescent device according to the related art, but gradually, phosphorescent material has been mainly used in the organic electroluminescent device. Therefore, in the electron transport material, which is a common material of the organic electroluminescent device, electron mobility appropriate for the phosphorescent material, low driving voltage, and a hole blocking property have been required.

Disclosure of Invention

Technical Problem

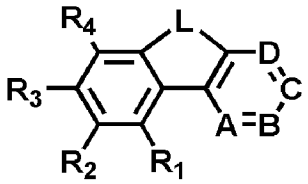
[10] An embodiment of the present invention is directed to providing a new electron transport material capable of significantly improving luminescence efficiency, stability and a lifespan of a device.

[11] In addition, an embodiment of the present invention is directed to providing an organic electroluminescent device capable of having an excellent luminescence property by using the new electron transport material and decreasing consumption power by decreasing driving voltage to induce increase in power efficiency.

Solution to Problem

[12] In one general aspect, there are provided an electron transport material represented by the following Chemical Formula 1, and an organic electroluminescent device containing the same. The electron transport material according to the present invention is used, such that excellent luminescence property may be obtained, and an increase in power efficiency is induced by decreasing driving voltage, such that the organic electroluminescent device using less consumption power may be manufactured.

[13] [Chemical Formula 1]

[14] 

[15] [In Chemical Formula 1,

[16] A, B, C, and D each are independently C-$(L_1)_m$-Ar_1 or N, but two of A, B, C, and D are N, the other two are C-$(L_1)_m$-Ar_1, and each of -$(L_1)_m$-Ar_1 may be the same as or different from each other, but two -$(L_1)_m$-Ar_1 are not hydrogen at the same time;

[17] L is (C1-C20)alkylene or (C2-C20)alkenylene, a carbon atom (-CH2-) of alkylene of L may be substituted with a heteroatom selected from NR(R is (C1-C30)alkyl), O, and S, and a carbon atom (=CH-) of alkenylene of L may be substituted with N;

[18] R_1 to R_4 each are independently hydrogen, (C1-C30)alkyl, (C3-C30)cycloalkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

[19] L_1(s) each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

[20] m is an integer of 1 to 3, and in the case in which m is an integer of 2 or more, L_1(s) each are the same as or different from each other;

[21] Ar_1(s) each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

[22] alkyl, cycloalkyl, aryl, heteroaryl of R_1 to R_4, alkylene or alkenylene of L, arylene and heteroarylene of L_1, and aryl and heteroaryl of Ar_1 may be substituted with at least one selected from a group consisting of (C1-C30)alkyl, halo(C1-C30)alkyl, halogen, cyano, (C3-C30)cycloalkyl, (C1-C30)alkoxy, (C6-C30)aryloxy, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, (C3-C30)heteroaryl, (C3-C30)heteroaryl substituted with (C1-C30)alkyl, (C3-C30)heteroaryl substituted

with (C6-C30)aryl, mono or di(C1-C30)alkylamino, mono or di(C6-C30)arylamino, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, nitro, and hydroxy; and

[23] Heteroarylene and heteroaryl contains at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

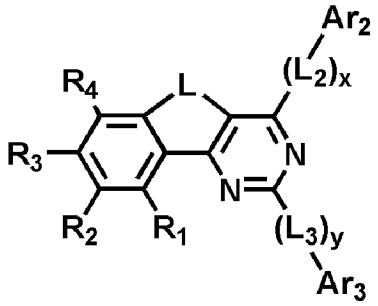
[24] The terms "alkyl", "alkoxy" and other substituents including an "alkyl" part described in the present invention include both of the straight chain type and the branched chain type, and the term "cycloalkyl" includes polycyclic hydrocarbon such as substituted or unsubstituted adamantyl or substituted or unsubstituted (C7-C30)bicycloalkyl as well as monocyclic ring system. The term "aryl" described herein, which is an organic radical derived from aromatic hydrocarbon by the removal of one hydrogen atom, may include a single ring or a fused ring containing, preferably 4 to 7 ring atoms, and more preferably 5 or 6 ring atoms, and include rings in which two or more aryl groups are combined through a single bond(s). Specific examples of aryl include phenyl, naphthyl, biphenyl, terphenyl, anthryl, indenyl, fluorenyl, phenanthryl, triphenylenyl, pyrenyl, perylenyl, chrysenyl, naphtacenyl, fluoranthenyl, and the like. The term "heteroaryl" described herein means an aryl group containing 1 to 4 hetero atom(s) selected from B, N, O, S, P(=O), Si, and P for the aromatic cyclic backbone atoms, and carbon atom(s) for remaining aromatic cyclic backbone atoms. The heteroaryl may be a 5- or 6-membered monocyclic heteroaryl or a polycyclic heteroaryl which is fused with one or more benzene ring(s), and may be partially saturated. In addition, the "heteroaryl"in the present invention may include the structures having one or more heteroaryl group(s) bonded through a single bond. The heteroaryl groups may include divalent aryl groups of which the heteroatoms are oxidized or quarternized, for example, to form N-oxides, quaternary salts, or the like. Specific examples of the heteroaryl group include monocyclic heteroaryl groups such as furyl, thiophenyl, pyrrolyl, imidazolyl, pyrazolyl, thiazolyl, thiadiazolyl, isothiazolyl, isoxazolyl, oxazolyl, oxadiazolyl, triazinyl, tetrazinyl, triazolyl, tetrazolyl, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, pyridazinyl, or the like; polycyclic heteroaryl groups such as benzofuranyl, benzothiophenyl, isobenzofuranyl, benzimidazolyl, benzothiazolyl, benzisothiazolyl, benzisoxazolyl, benzoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolinyl, quinazolinyl, quinoxalinyl, carbazolyl, phenanthridinyl, benzodioxolyl, dibenzofuranyl, dibenzothiophenyl, or the like; and corresponding N-oxides (for example, pyridyl N-oxide, quinolyl N-oxide); quaternary salts thereof, and the like.

[25] Further,'(C1-C30)alkyl'described herein may be preferably (C1-C20)alkyl, more preferably, (C1-C10)alkyl, and '(C6-C30)aryl'described herein may be preferably (C6-C20)aryl. '(C3-C30)heteroaryl'may be preferably (C3-C20)heteroaryl.

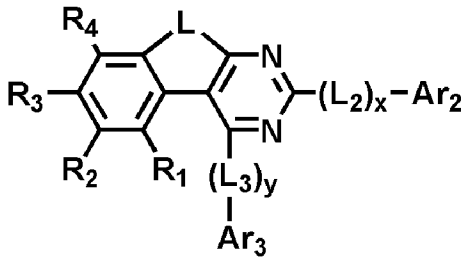
'(C3-C30)cycloalkyl'may be preferably (C3-C20)cycloalkyl, more preferably (C3-C7)cycloalkyl.

[26] More specifically, the electron transport material according to the present invention may be represented by the following Chemical Formula 2 or 3.

[27] [Chemical Formula 2]

[28] 

[29] [Chemical Formula 3]

[30] 

[31] [In Chemical Formulas 2 and 3,

[32] R_1 to R_4 and L each has the same definition in Chemical Formula 1;

[33] L_2 and L_3 each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

[34] x and y each are independently an integer of 1 to 3, wherein when x is an integer of 2 or more, L_2(s) are the same as or different from each other, and when y is an integer of 2 or more, L_3(s) are the same as or different from each other;

[35] Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

[36] Arylene and heteroarylene of L_2 and L_3, and aryl and heteroaryl of Ar_2 and Ar_3 may be substituted with at least one selected from a group consisting of (C1-C30)alkyl, halo(C1-C30)alkyl, halogen, cyano, (C3-C30)cycloalkyl, (C1-C30)alkoxy, (C6-C30)aryloxy, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, (C3-C30)heteroaryl, (C3-C30)heteroaryl substituted with (C1-C30)alkyl, (C3-C30)heteroaryl substituted with (C6-C30)aryl, mono or di(C1-C30)alkylamino, mono or di(C6-C30)arylamino, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, nitro, and hydroxy; and

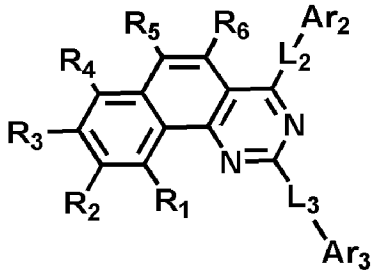
[37] $-(L_2)_x-Ar_2$ and $-(L_3)_y-Ar_3$ are not hydrogen at the same time.]

[38] More specifically, the electron transport material according to the present invention

may be represented by the following Chemical Formulas 4 to 9.

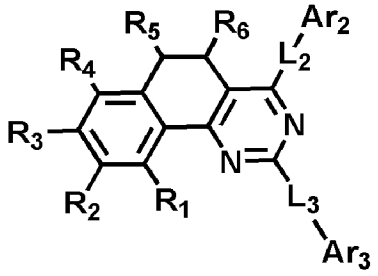
[39] [Chemical Formula 4]

[40]



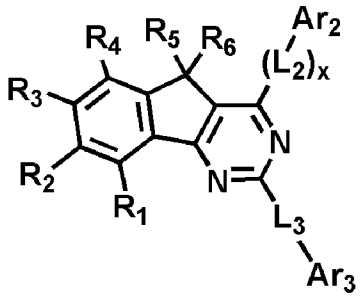
[41] [Chemical Formula 5]

[42]



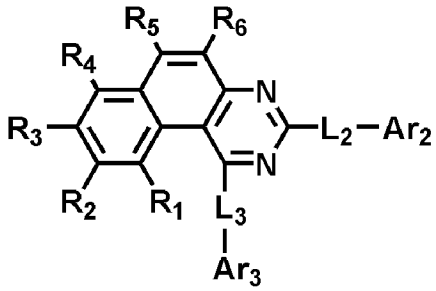
[43] [Chemical Formula 6]

[44]



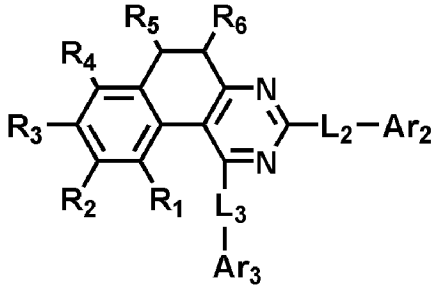
[45] [Chemical Formula 7]

[46]

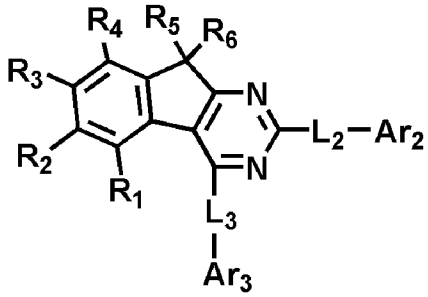


[47] [Chemical Formula 8]

[48]



[49] [Chemical Formula 9]

[50] 

[51] [In Chemical Formulas 4 to 9,

[52] R_1 to R_6 each are independently hydrogen, (C1-C30)alkyl, (C3-C30)cycloalkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

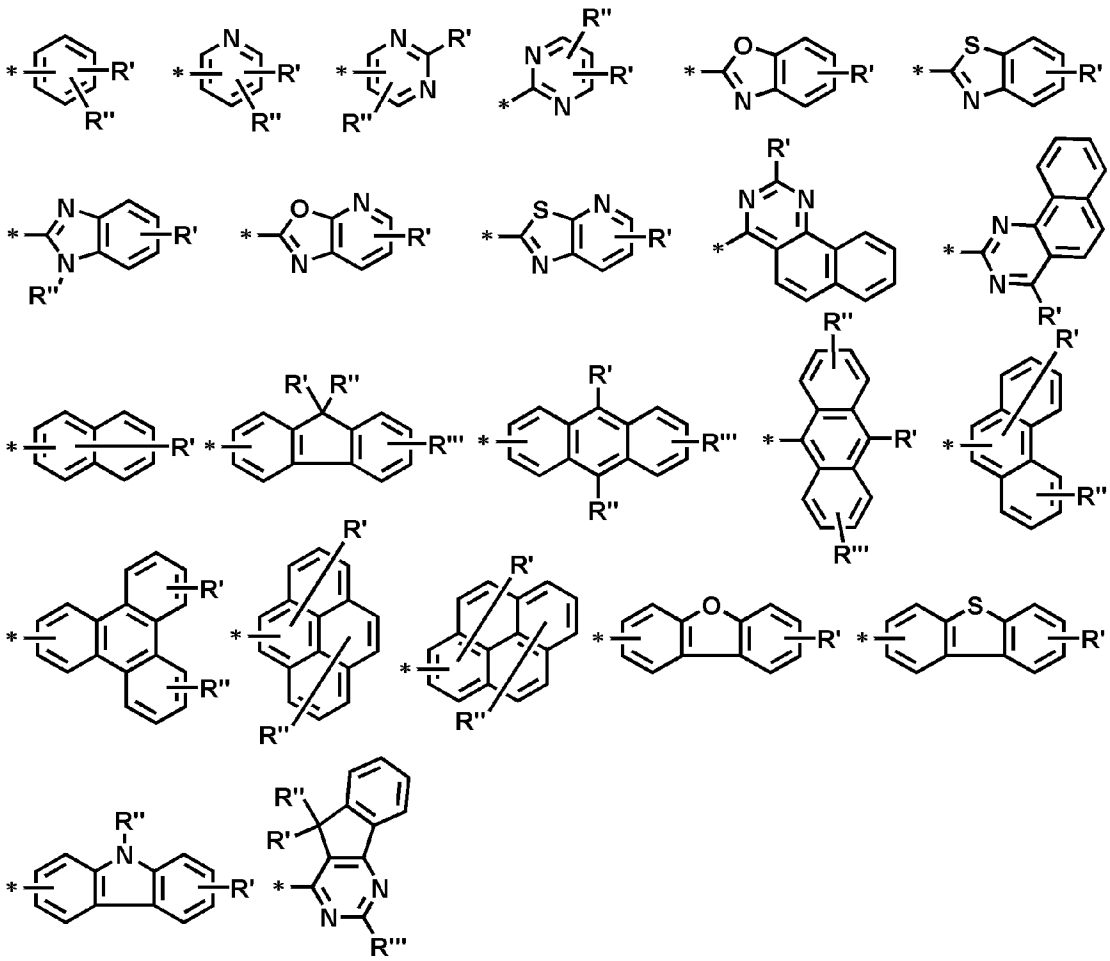
[53] L_2 and L_3 each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

[54] x is an integer of 1 to 3, and when x is an integer of 2 or more, L_1(s) each are the same as or different from each other,

[55] Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl; and arylene of L_2 and L_3 and aryl and heteroaryl of Ar_2 and Ar_3 may be further substituted with at least one selected from a group consisting of (C1-C30)alkyl, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, and (C3-C30)heteroaryl.]

[56] In the electron transport material of Chemical Formula 1, L_2 and L_3 each are a single bond, phenylene, biphenylene, 9,9-dimethylfluorenylene, naphthylene, anthrylene, pyridinylene, or pyrimidinylene; Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, or selected from the following structure; and

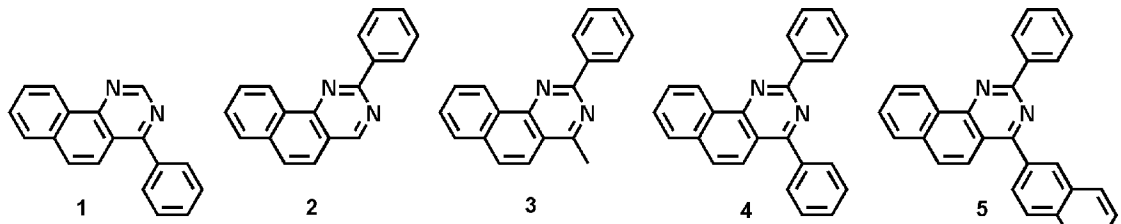
[57]



[58] R', R", and R'" each may be independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, (C3-C30)heteroaryl, or (C1-C30)alkyl(C6-C30)aryl.

[59] The electron transport material may be, for example, the following compounds, but is not limited thereto.

[60]



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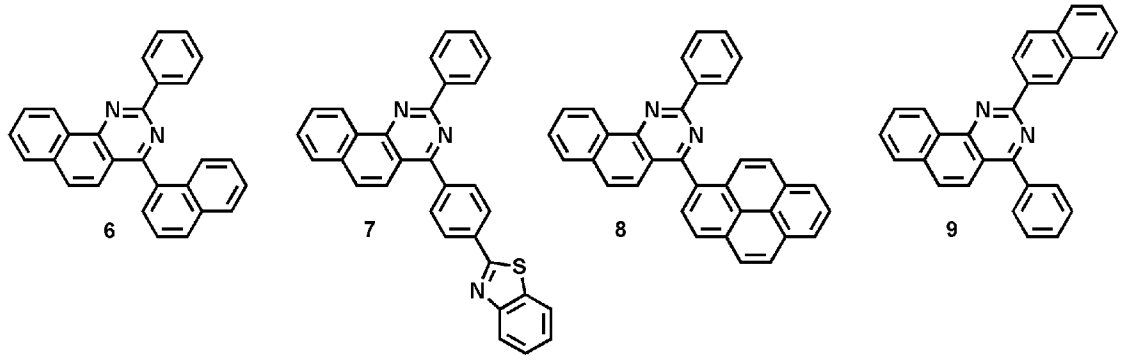

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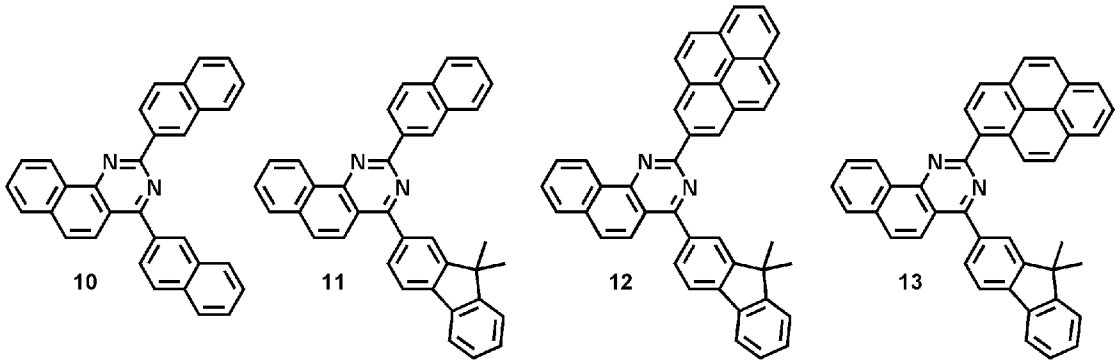

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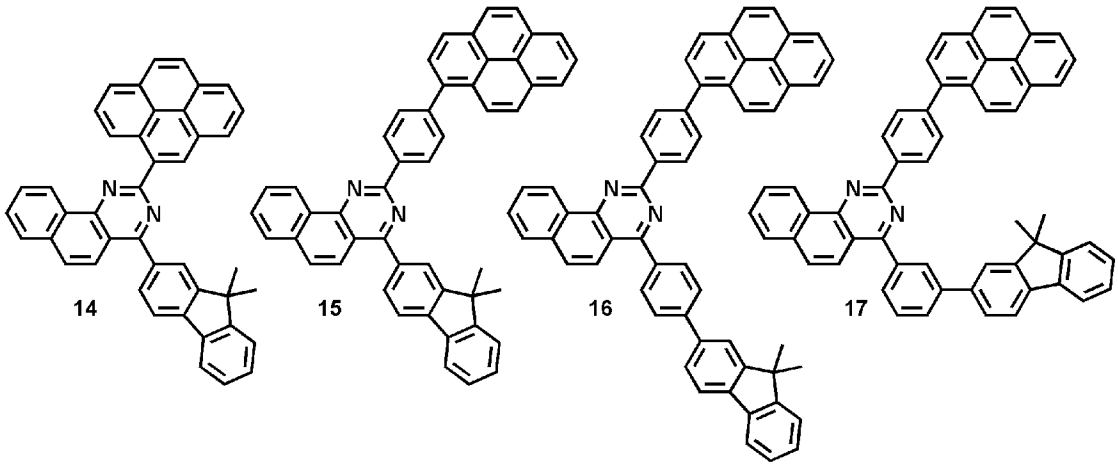

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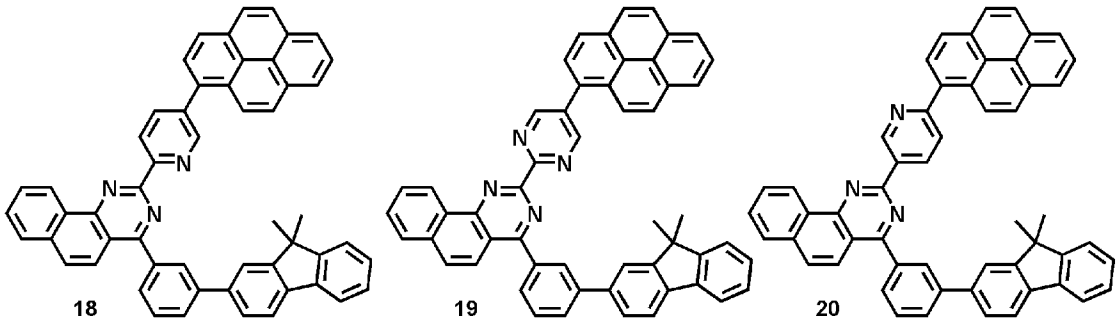

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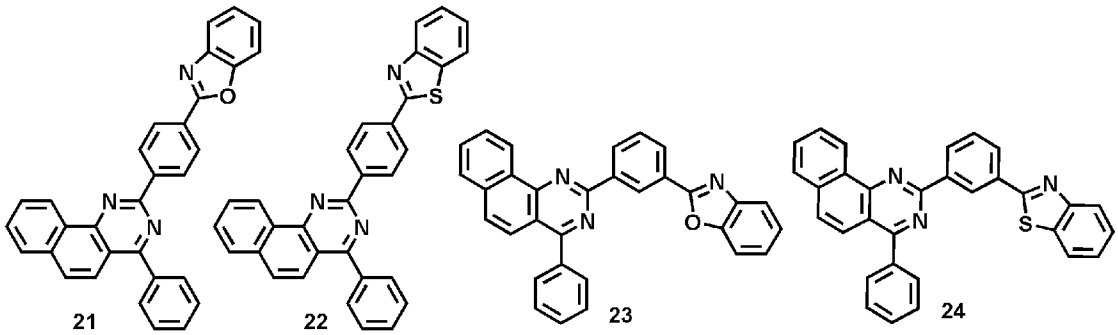

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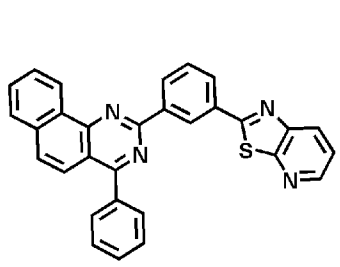
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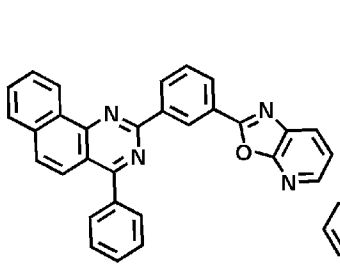
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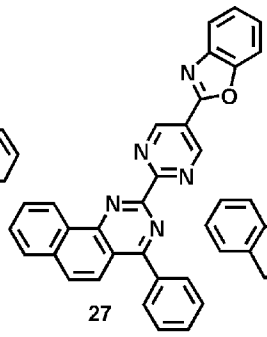
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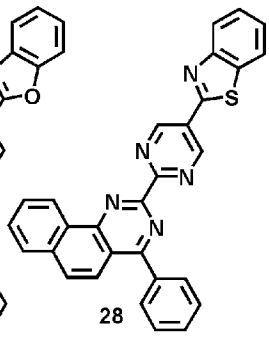
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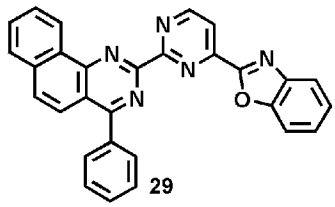
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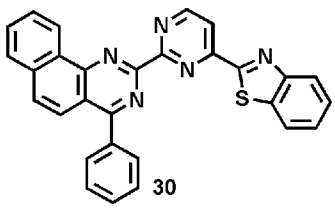
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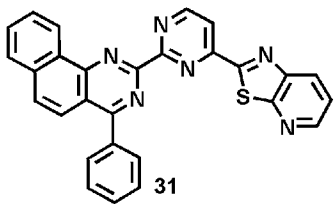
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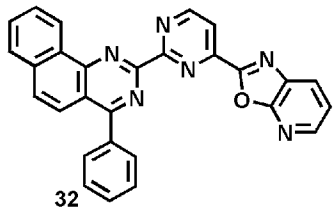
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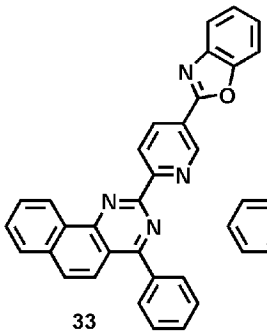
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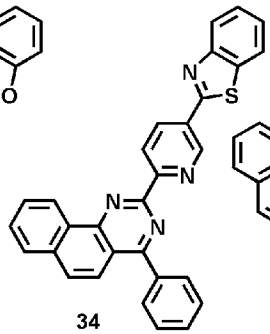
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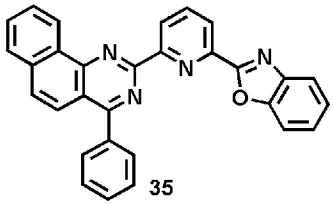
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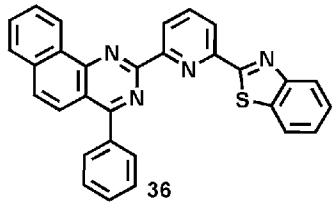
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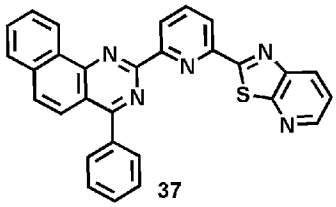
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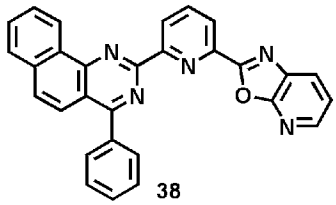
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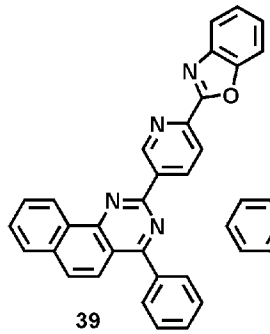
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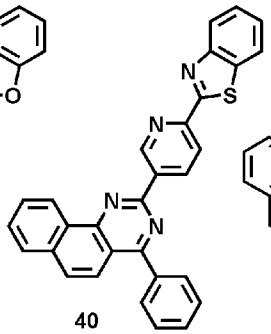
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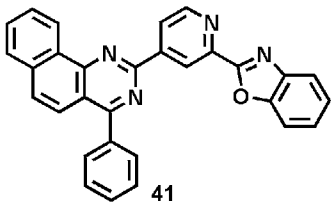
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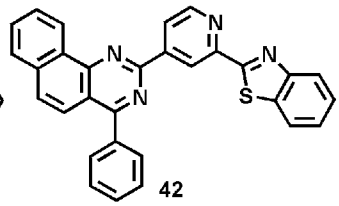
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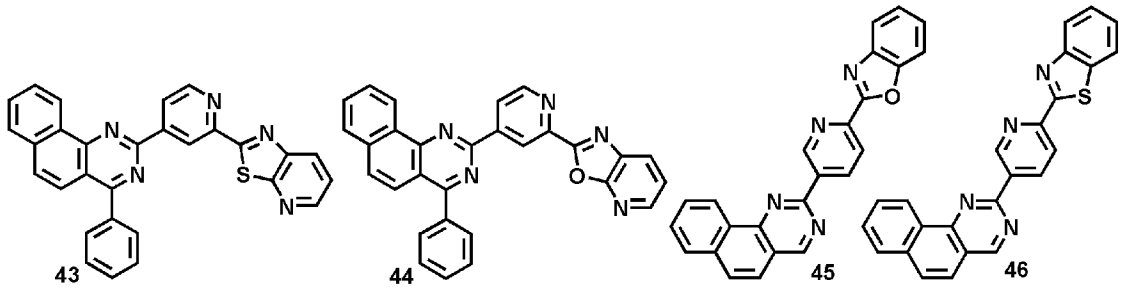

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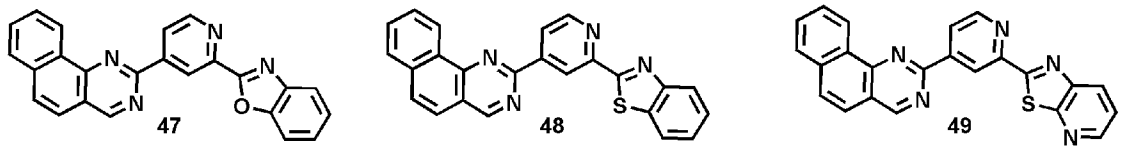

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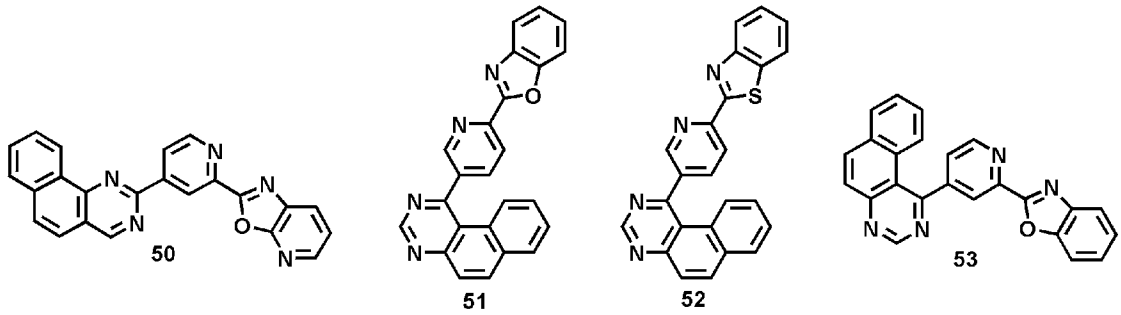

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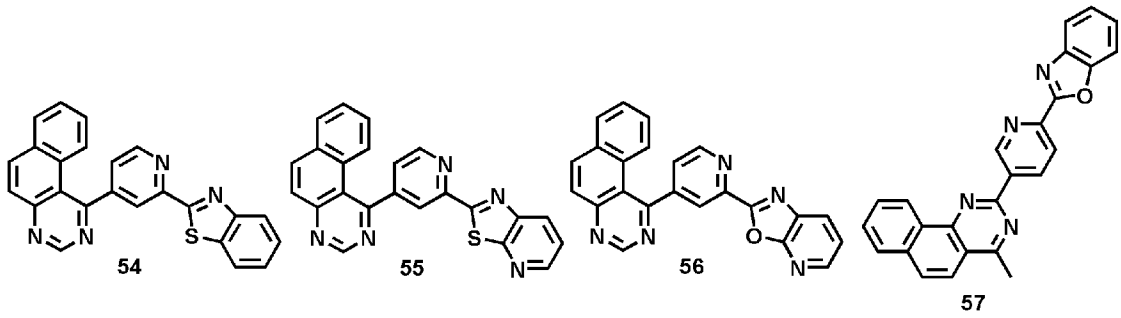

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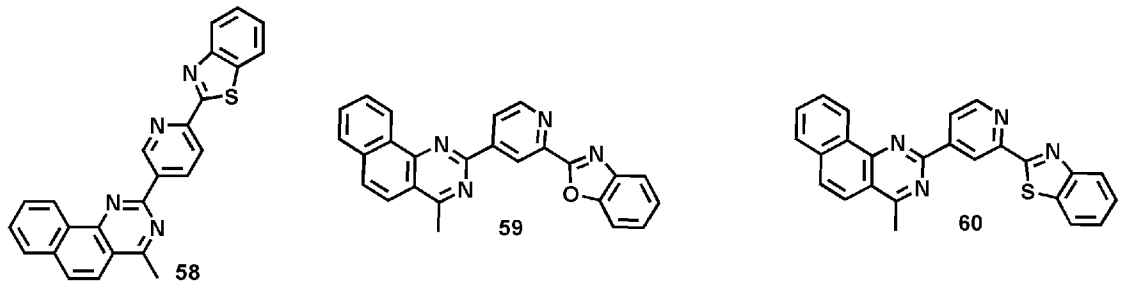

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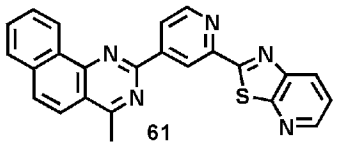

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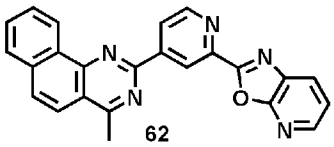

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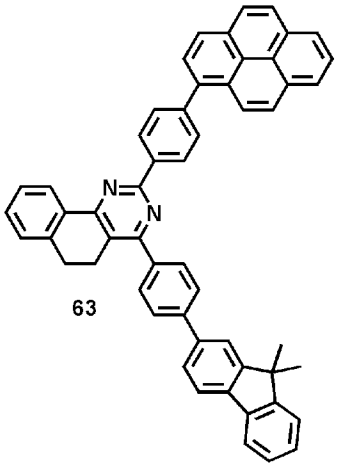

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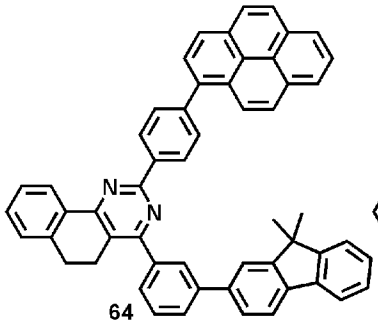

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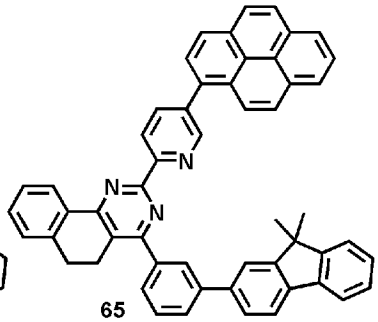

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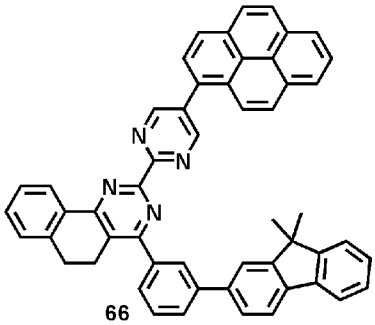

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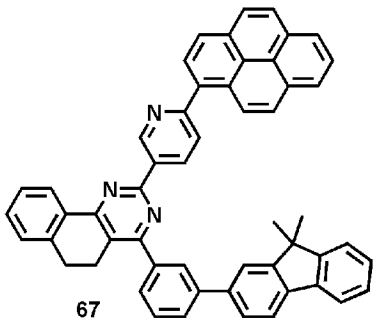

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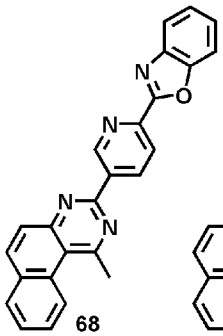

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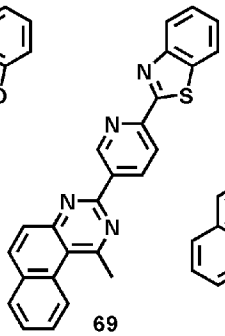

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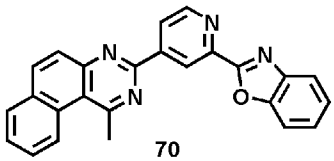

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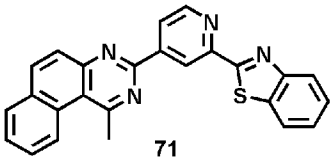

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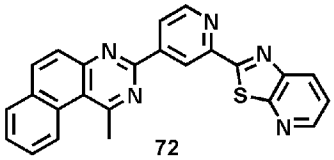

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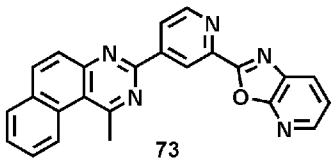

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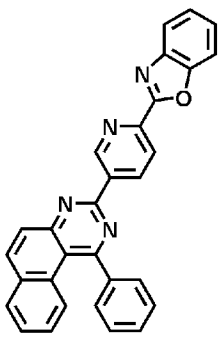

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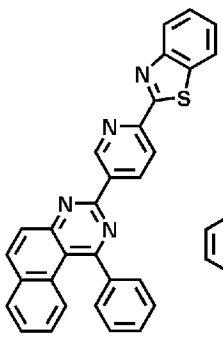

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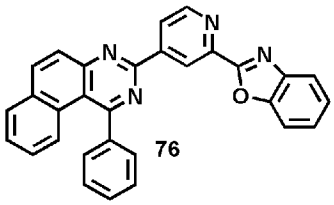

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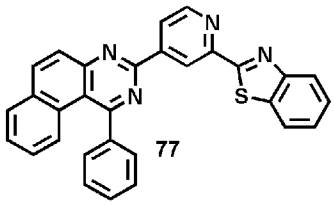

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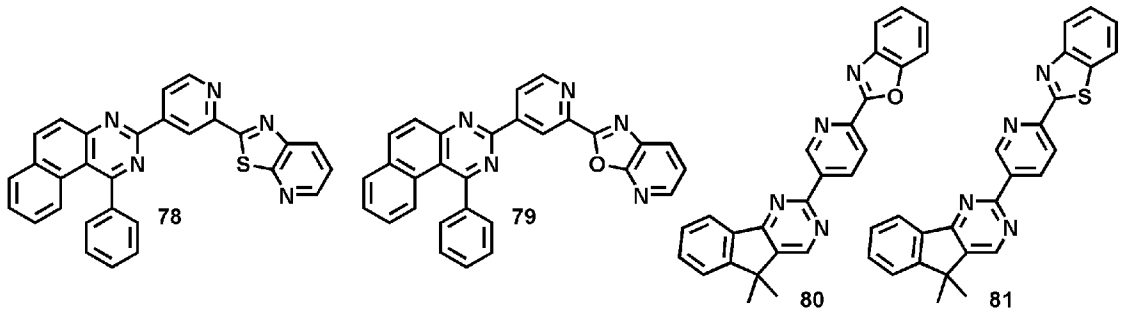

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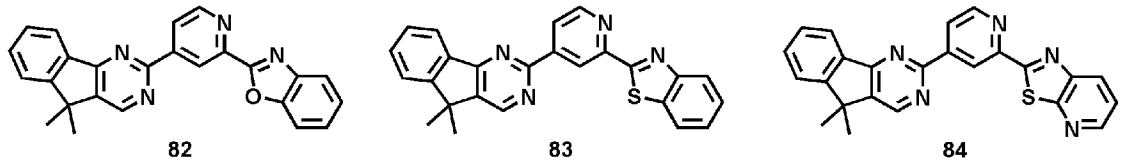

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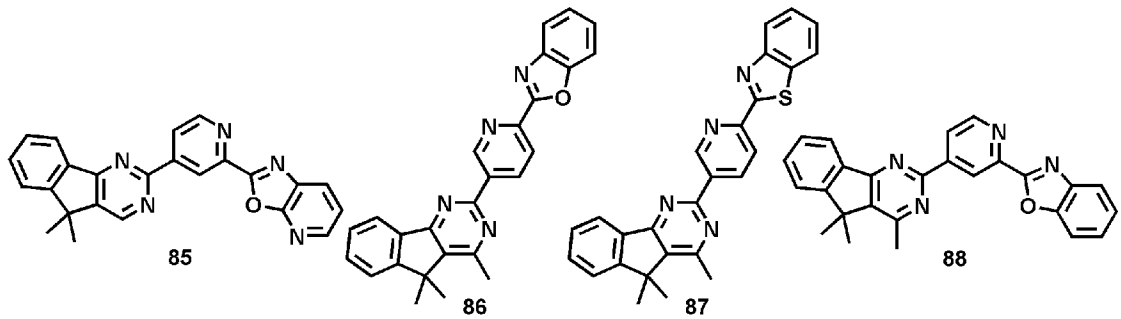

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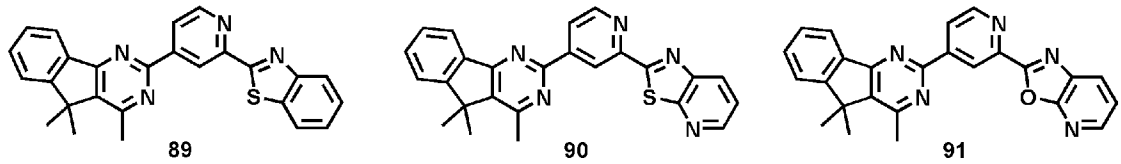

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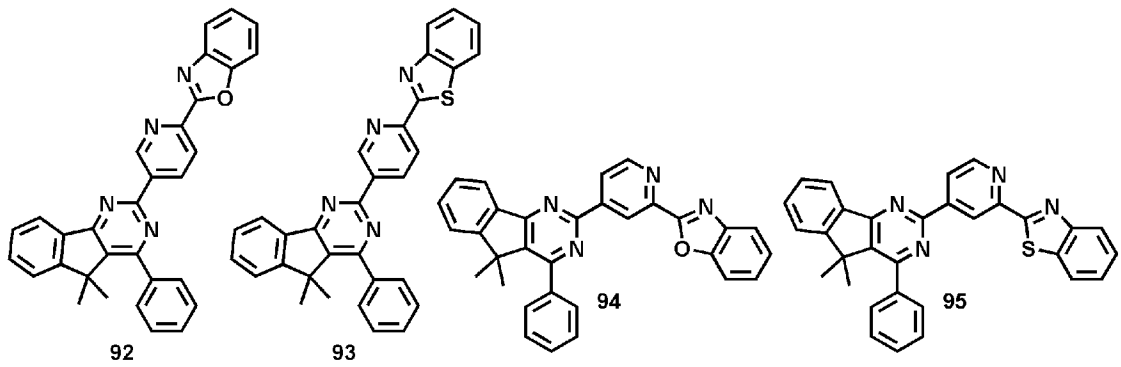

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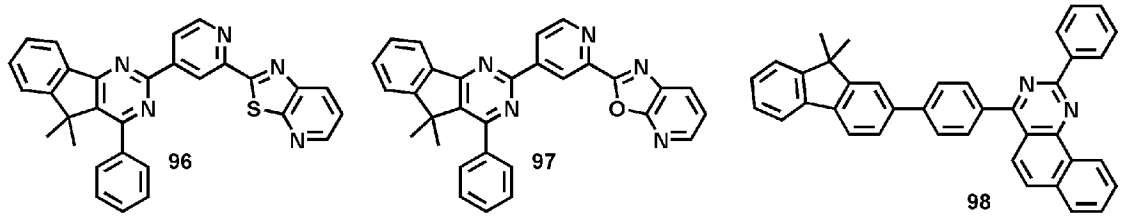

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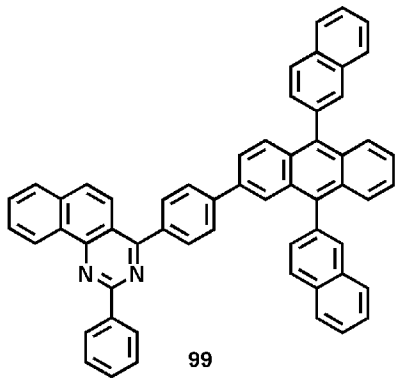

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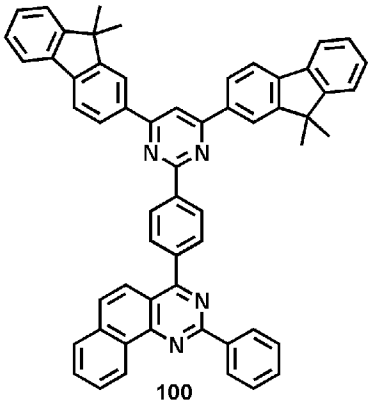

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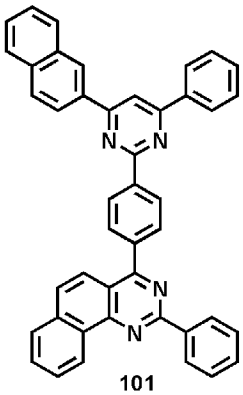

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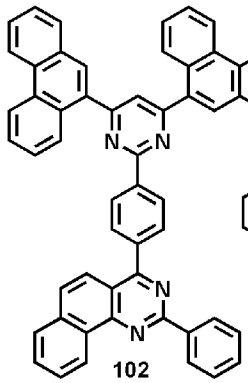

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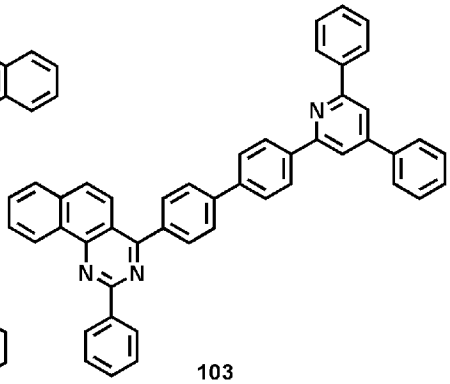

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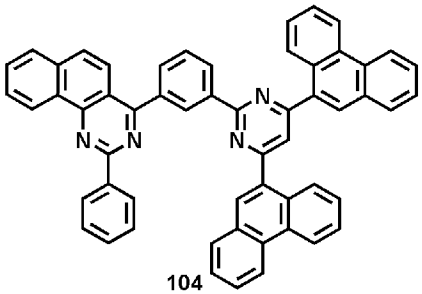

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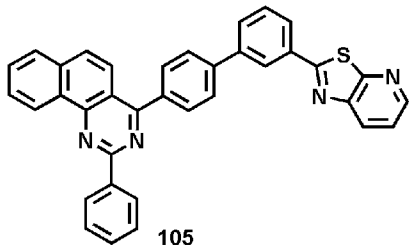

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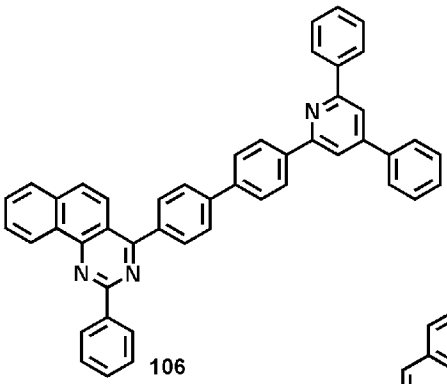

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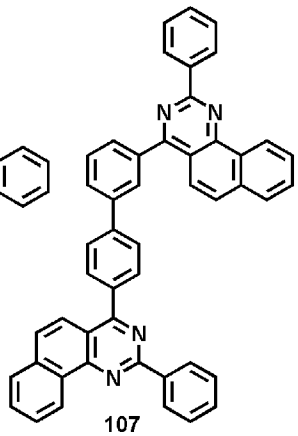

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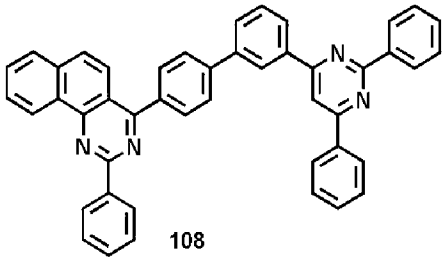

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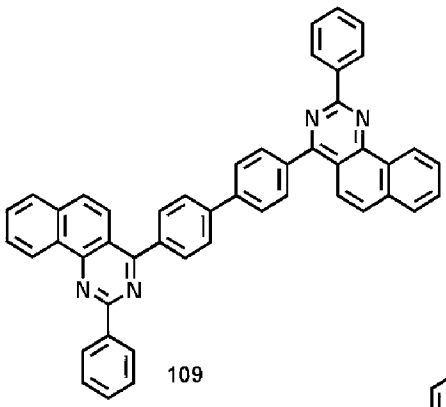

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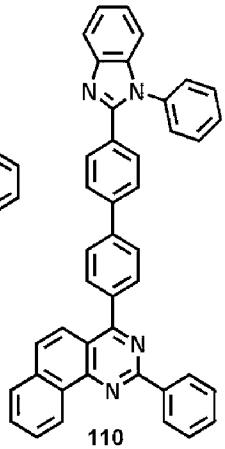

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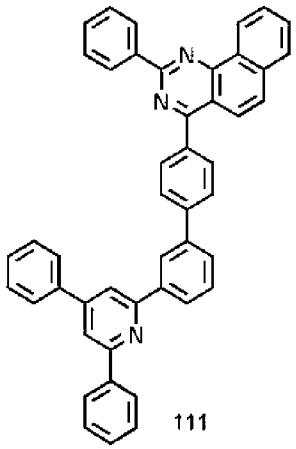

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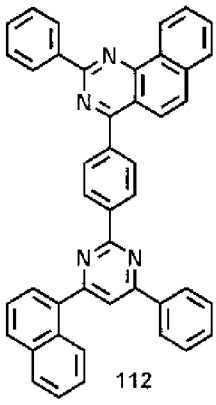

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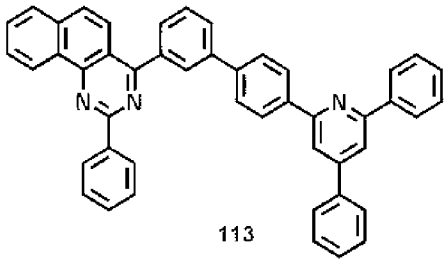

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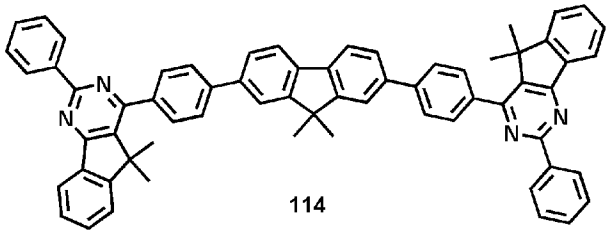

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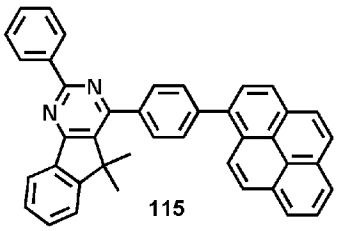

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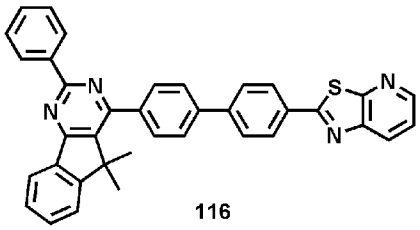
[94]



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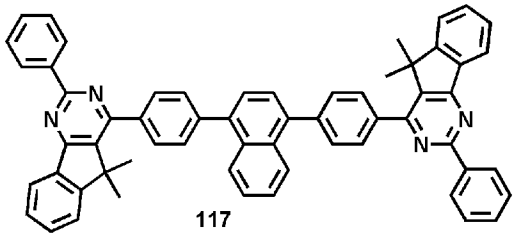

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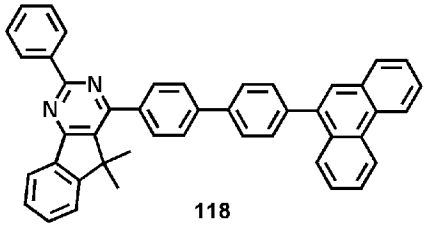

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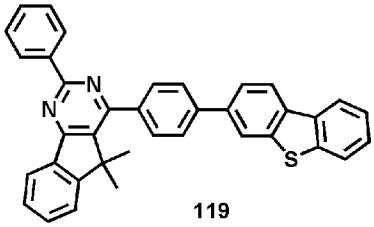

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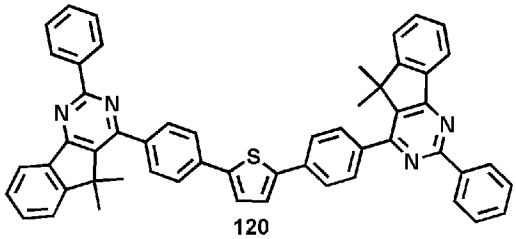

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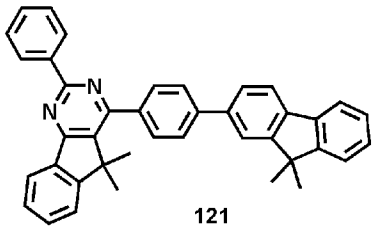

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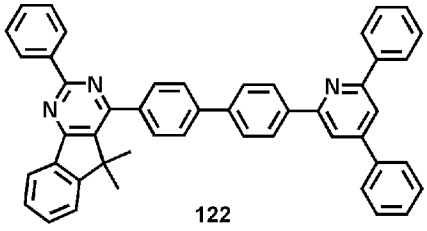

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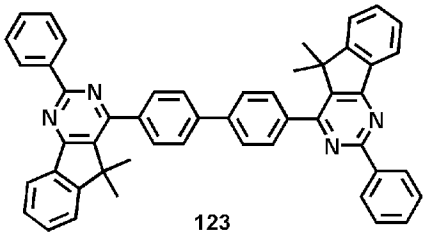

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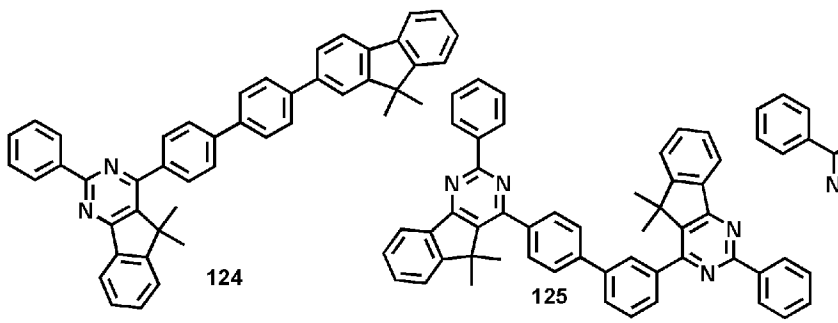
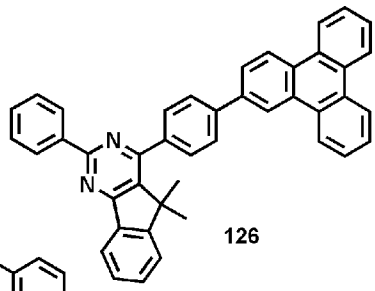
[98]





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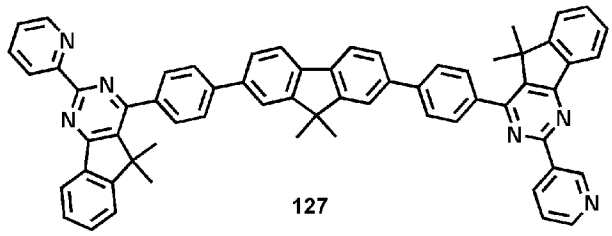

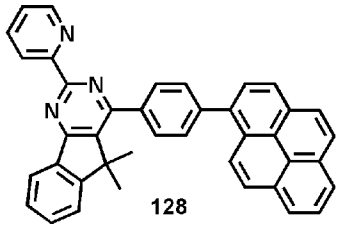

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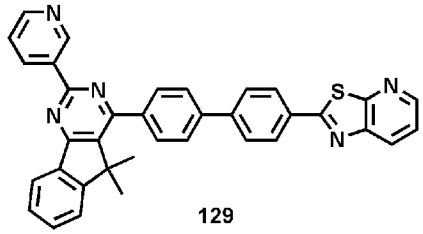

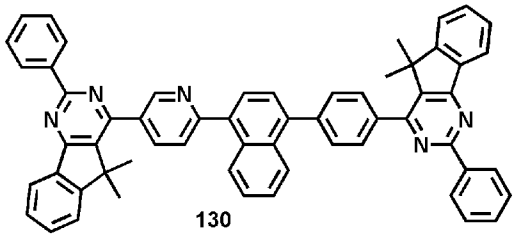

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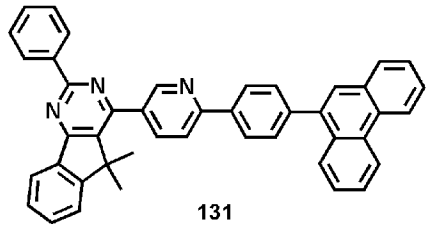

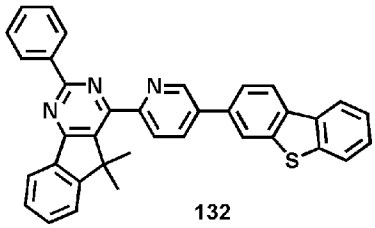

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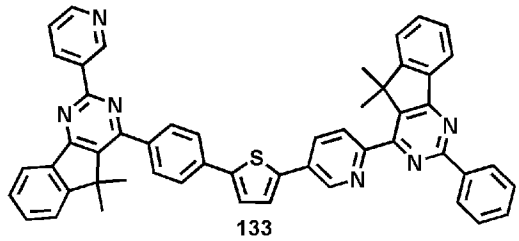

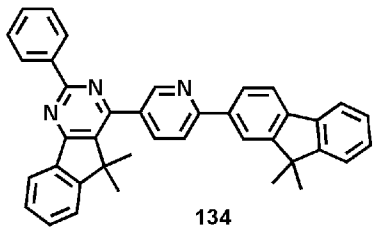

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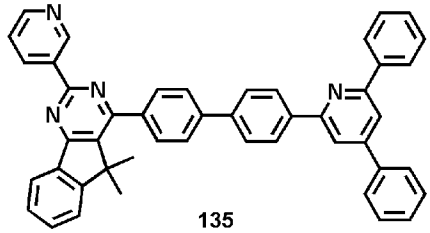
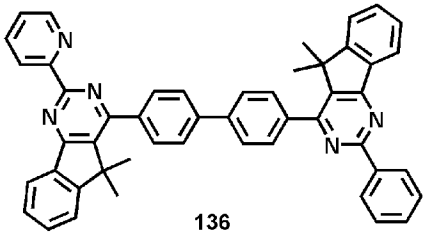
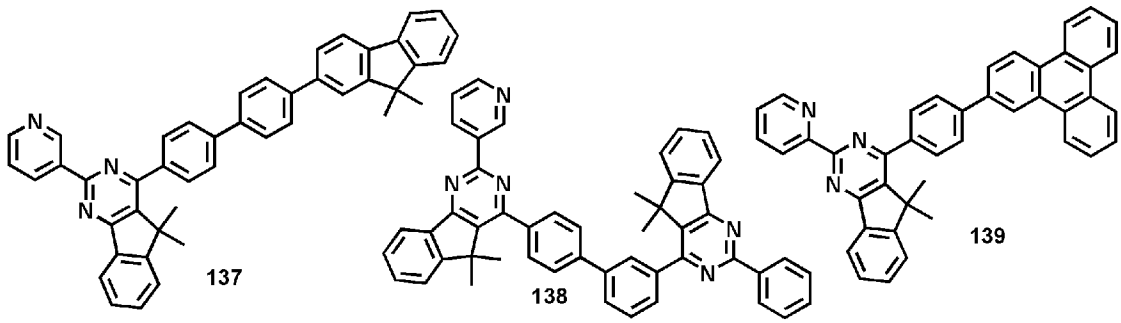




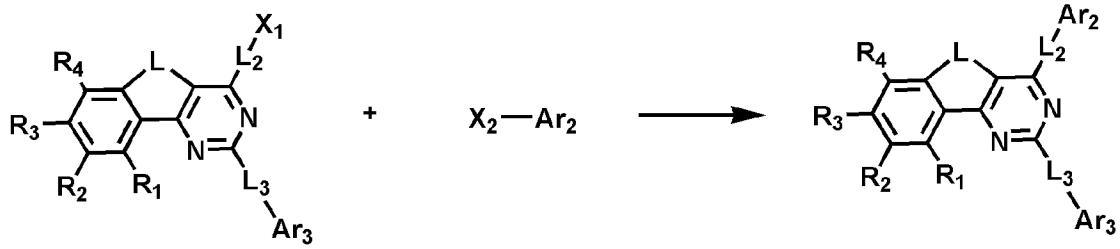
[104]

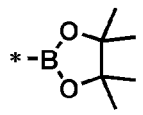


[105] Among the electron transport materials according to the present invention, a process of preparing an electron transport material of Chemical Formula 2 when x and y are 1 is represented by the following Reaction Formula 1, and a process of preparing an electron transport material of Chemical Formula 2 in the case in which x is 3 and y is 1 is represented by the following Reaction Formulas 2 to 4. However, The electron transport material is not limited thereto, but may be prepared by an organic reaction known in the art.

[106] [Reaction Formula 1]

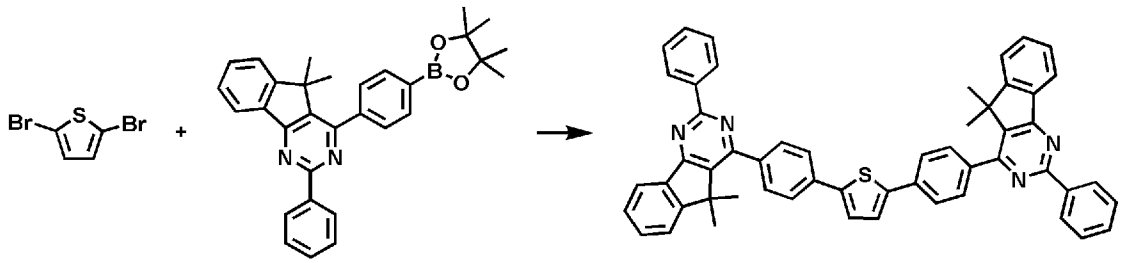
[107]



[108] [R_1 to R_4, L, L_2, L_3, Ar_2, and Ar_3 each has the same definition in Chemical Formula 2, X_1 and X_2 are halogen or  but are not equal to each other.]

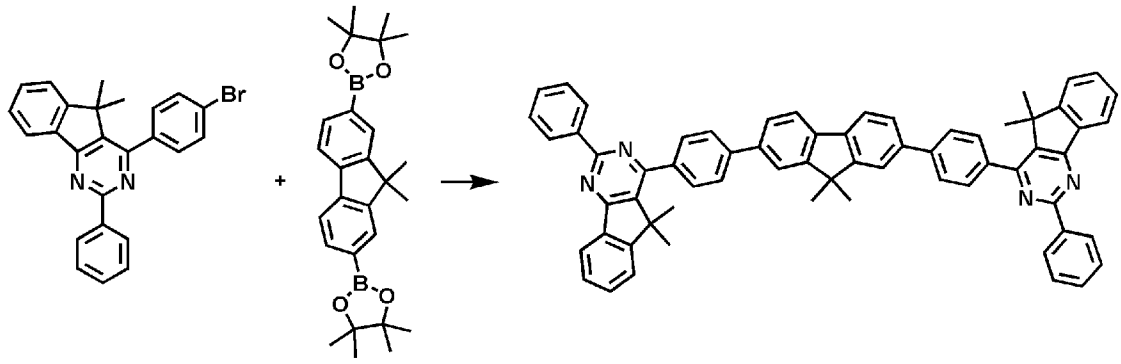
[109] [Reaction Formula 2]

[110]



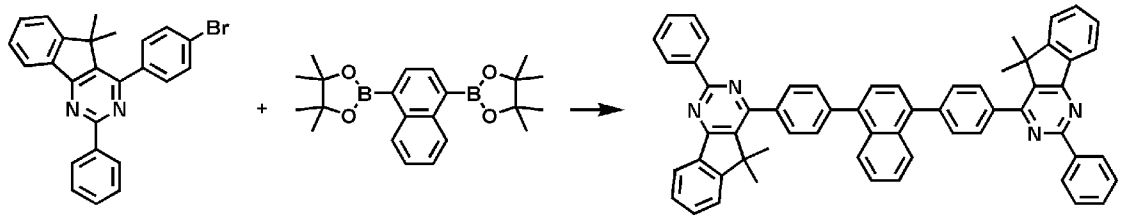
[111] [Reaction Formula 3]

[112]



[113] [Reaction Formula 4]

[114]



[115] In another general aspect, there is provided an organic electroluminescent device including a first electrode; a second electrode; and at least one organic layer interposed between the first and second electrodes, wherein the organic layer includes an electron transport layer containing an electron transport material of Chemical Formula 1. In the case in which the electron transport material of Chemical Formula 1 is used in the electron transport layer, driving voltage may be decreased, such that an increase in power efficiency may be induced, thereby decreasing consumption power.

[116] Further, the organic layer may include at least one electron transport layer in which the electron transport material of Chemical Formula 1 is contained and at least one luminescent layer configured of a fluorescent host and a fluorescent dopant or a phosphorescent host and a phosphorescent dopant, wherein the fluorescent host, the fluorescent dopant, the phosphorescent host, or the phosphorescent dopant are not particularly limited.

Advantageous Effects of Invention

[117] The electron transport material according to the present invention may have the excellent luminescence property and decrease the driving voltage to increase the power efficiency, such that the organic electroluminescent device using less consumption power may be manufactured.

Brief Description of Drawings

[118] FIG. 1 is a graph showing efficiency (cd/A) and luminance (cd/m^2) of organic electroluminescent devices manufactured in Examples 7 to 12 and Comparative Example 1.

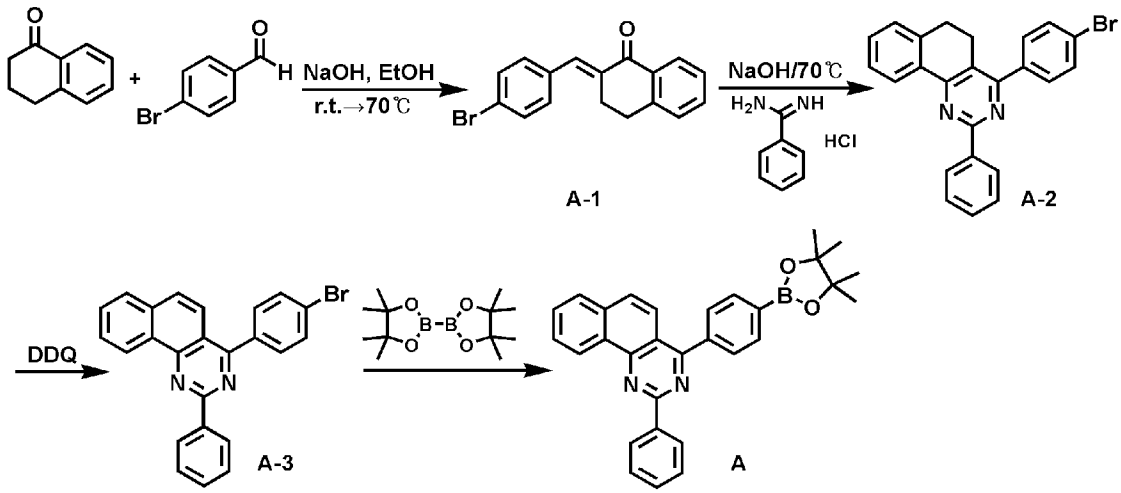
[119] FIG. 2 is a graph showing efficiency (cd/A) and luminance (cd/m^2) of organic electroluminescent devices manufactured in Examples 22 to 30 and Comparative Example

1

Mode for the Invention

[120] Hereinafter, an electron transport compound according to the present invention, a preparing method thereof, and luminescence properties of a device will be described in detail using a representative compound of the present invention as an example. However, the examples are for illustrating the present invention and not for limiting the present invention.

[121] [Preparation Example 1] Preparation of Compound A

[122] 

[123] Preparation of Compound A-1

[124] Alpha-tetralone (80g, 0.55mol) and 4-bromobenzaldehyde (106.5g, 0.58mol) were dissolved in ethanol (480mL), and then sodium hydroxide (27.4g, 0.68mol) was slowly added thereto at 0℃. After the mixture was stirred at room temperature for 3 hours, the prepared solid was separated by filtering under reduced pressure and sequentially washed with methanol, distilled water, and methanol. The washed material was dried, thereby obtaining Compound A-1 (148g, 69.8%) as a yellow solid.

[125] ^{1}H NMR ($CDCl_3$) d 7.97 (1H, d), 7.67-7.54 (7H, m), 7.46 (1H, t), 2.91 (2H, t), 2.73 (2H, t)

[126] Preparation of Compound A-2

[127] Compound A-1 (80g, 0.26mol), benzamidinehydrochloride (44g, 0.28mol), sodium hydroxide (15g, 0.38mol), and ethanol (1200mL) were mixed and refluxed for 12 hours. When the reaction was completed, the resultant material was cooled to room temperature, and the precipitated solid was filtered under reduced pressure and washed with water and ethanol. The washed material was dried, thereby obtaining Compound A-2 (50g, 47.4%) as a white solid.

[128] ^{1}H NMR ($CDCl_3$) d 8.65 (3H, m), 7.70 (4H, q), 7.51 (5H, m), 7.28 (1H, s), 3.09 (2H, t), 2.93 (2H, t)

[129] Preparation of Compound A-3

[130] Compound A-2 (20g, 0.048mol), 2,3-dichloro-5,6-dicyanoparabenzoquinone (DDQ, 27.5g, 0.12mol), and dichlorobenzene (200mL) were mixed and stirred at 120 for 4 hours. When the reaction was completed, the resultant material was cooled to room temperature and dispersed in methanol, followed by filtering under reduced pressure, thereby obtaining Compound A-3 (13.2g, 66.3%) as a white solid.

[131] ^{1}H NMR ($CDCl_3$) d 8.65 (3H, m), 7.70 (6H, q), 7.51 (7H, m), 7.28 (1H, s)

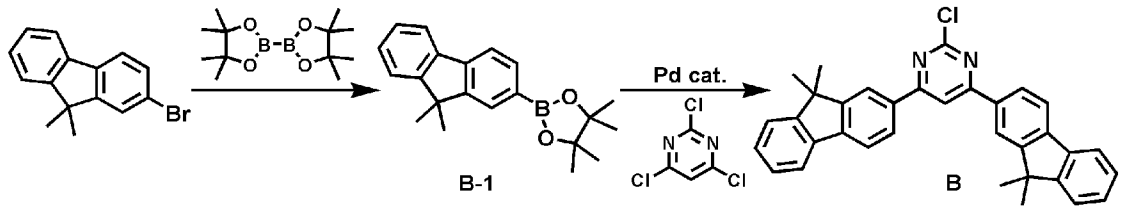
[132] Preparation of Compound A

[133] Compound A-3(10g, 24.3mmol), bis(pinacolato)diboron (6.48g, 25.5mmol), 1,4-dioxane (200mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (0.36g, 0.49mmol), and potassium acetate (4.8g, 48.6mmol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound A (9.8g, 87.9%) as a yellow solid.

[134] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.68 (6H, q), 7.55 (7H, m), 7.12 (1H, s), 1.40 (s, 12H)

[135]

[136] [Preparation Example 2] Preparation of Compound B

[137] 

[138] Preparation of Compound B-1

[139] 2-bromo-9,9-dimethylfluorene (10g, 24.3mmol), bis(pinacolato)diboron (19.52g, 76.88mmol), 1,4-dioxane (400mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (1.07g, 1.46mmol), and potassium acetate (14.37g, 146.43mmol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound B-1 (18g, 76.8%) as a white solid.

[140] ^{1}H NMR ($CDCl_3$) d 7.78 (2H, d), 7.57 (2H, m), 7.38 (3H, m), 1.58 (s, 6H), 1.40 (s, 12H)

[141] Preparation of Compound B

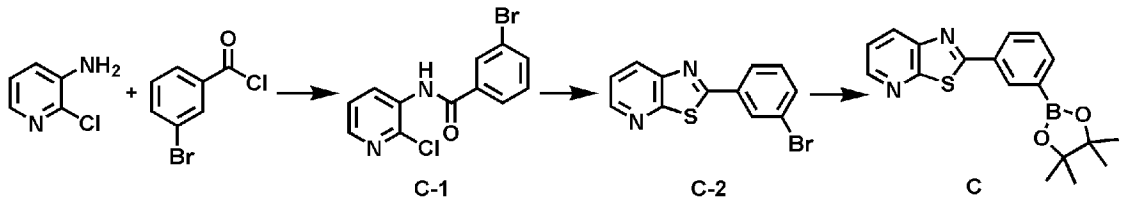
[142] Trichloropyrimidine (5g, 27.26mmol), Compound B-1(17.6g, 55.1mmol), $Pd(PPh_3)_4$ (0.63g, 0.55mmol), K_2CO_3(7.53g, 54.52mmol), distilled water (25mL), and THF(50mL) were mixed and stirred under reflux for 12 hours. After an organic layer was extracted with MC, the extracted organic layer was concentrated under reduced pressure and washed with acetone, thereby obtaining Compound B (9.3g, 68.4%).

[143] ^{1}H NMR ($CDCl_3$) d 8.29 (d, 2H), 8.18-8.16 (d, 2H), 8.12 (s, 1H), 7.9 (d, 2H), 7.84-7.82 (m, 2H), 7.53-7.51 (m, 2H), 7.44-7.41 (m, 4H), 1.62 (s, 12H)

[144]

[145] [Preparation Example 3] Preparation of Compound C

[146]



[147] Preparation of Compound C-1

[148] Dichloromethane (31mL) and 3-bromobenzoyl chloride (25g, 0.1139mol) were mixed and stirred at -10℃. After 3-bromobenzoyl chloride was completely dissolved, a mixing solution of pyridine (106mL) and 2-chloropyridine-3-amine (13.2g, 0.1026mol) was slowly dropped thereinto and stirred. After 2 hours 30 minutes, water (1250mL) was added thereto, and the prepared solid compound was filtered and washed with methanol. The resultant material was dried, thereby obtaining Compound C-1(28.0g, 87%) as a white solid.

[149] ^{1}H NMR ($CDCl_3$) d 8.58 (m, 1H), 8.31 (d, 1H), 8.30 (t, 1H), 8.14 (m, 1H), 7.90 (m, 1H), 7.45 (q, 1H), 7.40 (t, 1H)

[150] Preparation of Compound C-2

[151] 1,3-dimethyl-3,4,5,6-tetrahydro-2(1H)-pyrimidinone (DMPU, 110mL), Compound C-1 (27g, 0.0866mol), and Lawesson`s reagent (35.1g) were mixed and stirred under reflux. After 2 hours 30 minutes, the mixture was extracted with ethyl acetate (EA) and concentrated, thereby obtaining Compound C-2 (20g, 80%) as a light yellow solid.

[152] ^{1}H NMR ($CDCl_3$) d 8.62-8.60 (d, 1H), 8.33 (d, 1H), 8.30 (t, 1H), 8.03-8.00 (d, 1H), 7.68-7.65 (d, 1H), 7.50-7.46 (q, 1H), 7.41 (t, 1H)

[153] Preparation of Compound C

[154] Compound C-2 (27.47g, 103mmol), bis(pinacolato)diboron (27.47g, 108mmol), 1,4-dioxane (450mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (1.68g, 2.1mmol), and potassium acetate (20.22g,

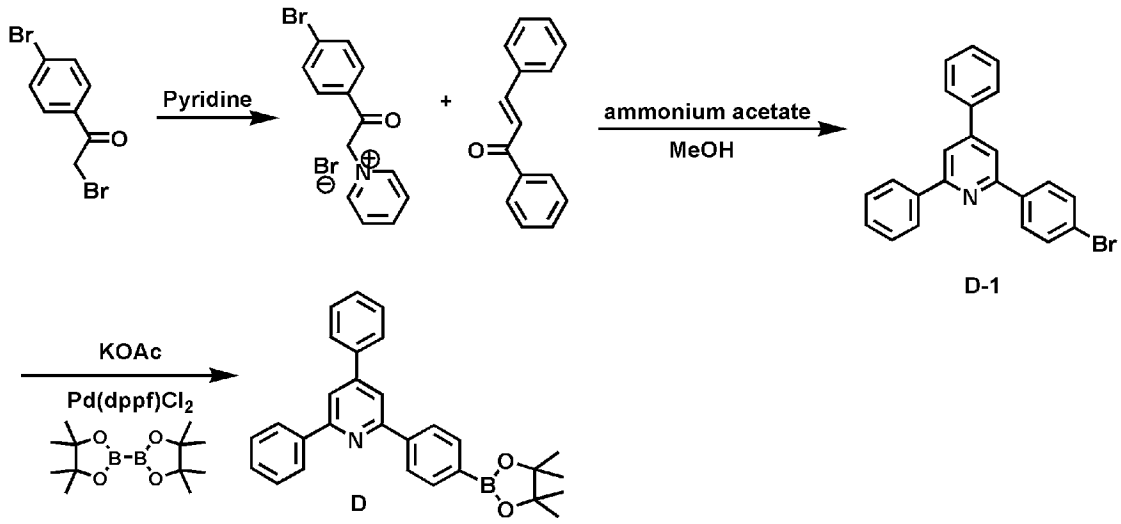
206.1mmol) were mixed and stirred under reflux for 12 hours. Then, the mixture was cooled to room temperature. An organic layer was extracted by adding saturated sodium chloride (NaCl) aqueous solution and ethyl acetate (EA) to the reactant, dried over magnesium sulfate ($MgSO_4$), and treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended and stirred in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound C (41.8g, 89 %).

[155] ^{1}H NMR ($CDCl_3$) d 8.60-8.58 (d, 1H), 8.50 (s, 1H), 8.32-8.30 (d, 1H), 8.25-8.22 (d, 1H), 7.99-7.97 (d, 1H), 7.55 (t, 1H), 7.48-7.44 (q, 1H), 1.40 (s, 12H)

[156]

[157] [Preparation Example 4] Preparation of Compound D

[158]



[159] Preparation of Compound D-1

[160] 4-bromophenacyl bromide (100g, 360mmol) was slowly added to pyridine (1000mL) while stirring pyridine (1000mL). After the mixture was stirred at room temperature for 2 hours, the precipitated solid was separated by filtering under reduced pressure and washed with methanol. The prepared compound (120g, 336mmol), trans-Chalcone (35g, 168mmol), ammonium acetate (25.9g, 336mmol), and methanol (450mL) were mixed and stirred under reflux for 12 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was separated by filtering under reduced pressure and washed with methanol, thereby obtaining Compound D-1 (25.5g, 47%) as a white solid.

[161] ^{1}H NMR ($CDCl_3$) d 7.46-7.58(m, 6H), 7.65(d, 2H), 7.76(d, 2H), 7.78(d, 2H), 8.12(d, 2H), 8.22(d, 2H)

[162] Preparation of Compound D

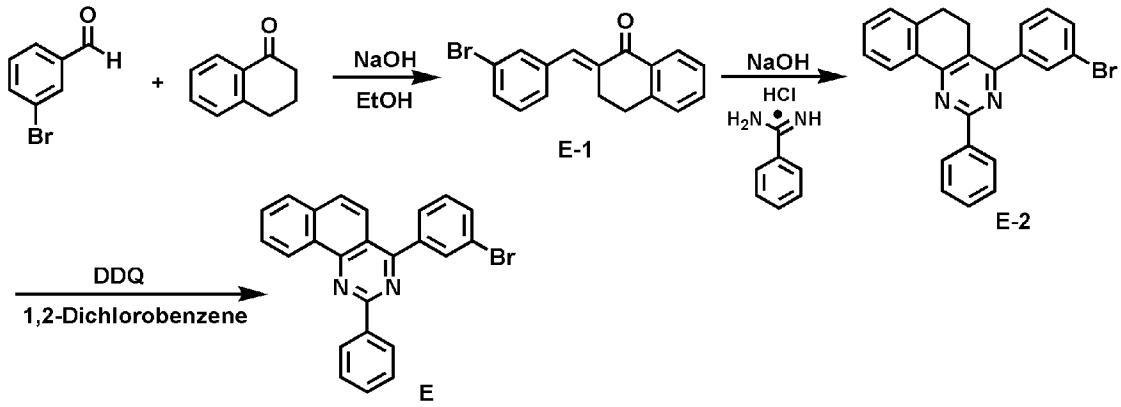
[163] Compound D-1 (25g, 64.7mmol), bis(pinacolato)diboron, (19.7g, 77.6mmol), 1,4-dioxane (600mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride

dichloromethane complex (1.6g, 1.9mmol), and potassium acetate (12.7g, 129mmol) were mixed and stirred under reflux for 12 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered under reduced pressure and separated using a silica gel column (hexane: ethyl acetate =10:1), thereby obtaining Compound D (21.6g, 77%) as a white solid.

[164] ^{1}H NMR ($CDCl_3$) d 1.41(s, 12H), 7.47-7.58(m, 6H), 7.77(d, 2H), 7.92-7.99(m, 4H), 8.21-8.25(m, 4H)

[165]

[166] [Preparation Example 5] Preparation of Compound E

[167] 

[168] Preparation of Compound E-1

[169] 3-bromobenzaldehyde (35g, 239mmol) and 1-tetralone (46.6g, 252mmol) were dissolved in ethanol (200mL), and then sodium hydroxide (11.9g, 299mmol) was slowly added thereto at 0°C. After the mixture was stirred at room temperature for 12 hours, the prepared solid was separated by filtering under reduced pressure and washed with methanol, thereby obtaining Compound E-1 (64.2g, 85%) as a light yellow solid.

[170] ^{1}H NMR ($CDCl_3$) d 2.98(t, 2H), 3.12(t, 2H), 7.27-7.41(m, 4H), 7.49-7.54(m, 2H), 7.59(s, 1H), 7.79(s, 1H), 8.14(d, 1H)

[171] Preparation of Compound E-2

[172] Compound E-1 (60g, 190mmol), benzamidinehydrochloride (33g, 210mmol), sodium hydroxide (11.4g, 290mmol), and ethanol (600mL) were mixed and refluxed for 12 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered under reduced pressure and separated using a silica gel column (hexane: methylene chloride =1:1), thereby obtaining Compound E-2 (54.4g, 68%) as a light yellow solid.

[173] ^{1}H NMR ($CDCl_3$) d 2.87(t, 2H), 3.02(t, 2H), 7.24(s, 1H), 7.34-7.61(m, 7H), 7.87(s, 2H), 8.56-8.63(m, 3H)

[174] Preparation of Compound E

[175] Compound E-2 (62.7g, 151mmol), DDQ (34g, 151mmol), and 1,2-dichlorobenzene (600mL) were mixed and refluxed at 120°C for 12 hours. When the reaction was

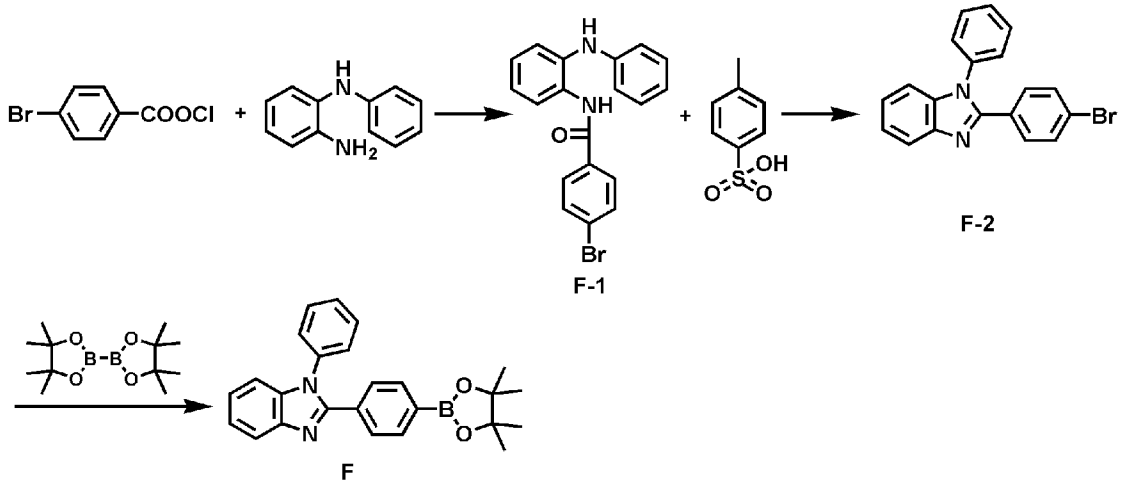
completed, the reactant was cooled to room temperature and extracted with water and methylene chloride, followed by separation using a silica gel column (hexane: methylene chloride =1:1), thereby obtaining Compound E (22.42g, 36%) as a light yellow solid.

[176] ^{1}H NMR ($CDCl_3$) d 7.47-7.59(m, 4H), 7.72-8.07(m, 8H), 8.82(d, 2H), 9.54(m, 1H)

[177]

[178] [Preparation Example 6] Preparation of Compound F

[179]



[180] Preparation of Compound F-1

[181] N1-phenylbenzene-1,2-diamine (48g, 260mmol) was completely dissolved in N,N-Dimethyl acetamide (DMAC, 100mL) at 0℃, and then 4-bromobenzoyl chloride (63g, 287mmol) was dropped thereinto and stirred. After 2 hours 30 minutes, pyridine (60mL) and water (100mL) were added thereto. After stirring the mixture for 30 minutes, the prepared solid was filtered while being washed with methanol, thereby obtaining Compound F-1 (95g, 95%) as a white solid.

[182] Preparation of Compound F-2

[183] Compound F-1 (50g, 136mmol), p-toluene sulfonic acid (PTSA, 46.9g, 272mmol), and toluene (500mL) were mixed and stirred under reflux with a Dean-stark trap under nitrogen atmosphere. After 2 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid obtained by concentrating the filtrate under reduced pressure was re-crystallized, thereby obtaining Compound F-2 (39g, 81%).

[184] ^{1}H NMR ($CDCl_3$) d 8.24(d, 1H), 7.66(m, 3H), 7.62-7.46(m, 6H), 7.40(m, 2H), 7.32(d, 2H)

[185] Preparation of Compound F

[186] Compound F-2 (38.6g, 110mmol), potassium acetate (32.4g, 330mmol),

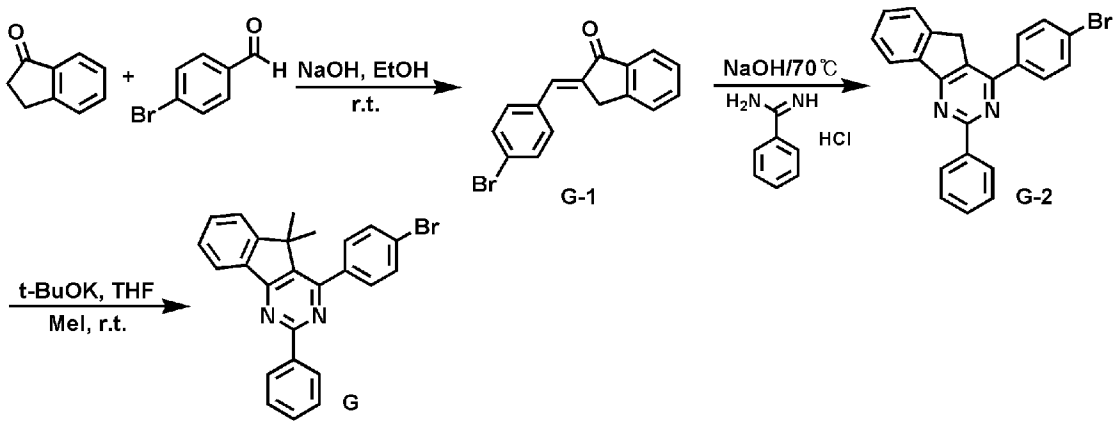
bis(pinacolato)diboron (36.5g, 143mmol), 1,4-dioxane (390mL), and $PdCl_2$(dppf) (1.8g, 2mmol) were mixed and stirred at 80 for 18 hours, and then cooled at room temperature. Water (400mL) was added to the reactant and stirred. After stirring, an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, dried over magnesium sulfate, and treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was re-crystallized, thereby obtaining Compound F (36g, 82%).

[187] ^{1}H NMR ($CDCl_3$) d 7.92(d, 1H), 7.76(d, 2H), 7.59(d, 2H), 7.56-7.45(m, 3H), 7.41-7.29(m, 4H), 7.27(s, 1H), 1.35(s, 12H)

[188]

[189] [Preparation Example 7] Preparation of Compound G

[190]



[191] Preparation of Compound G-1

[192] 1-indanone (80g, 0.61mol) and 4-bromobenzaldehyde (117.8g, 0.64mol) were dissolved in ethanol (1280mL), and then sodium hydroxide (30.3g, 0.76mol) was slowly added thereto at 0°C. After the mixture was stirred at room temperature for 3 hours, the prepared solid was separated by filtering under reduced pressure and sequentially washed with methanol, distilled water, and methanol. The resultant material was dried, thereby obtaining Compound G-1 (130g, 71.8%) as a yellow solid.

[193] ^{1}H NMR ($CDCl_3$) d 7.97 (1H, d), 7.67-7.54 (7H, m), 7.46 (1H, t), 3.32 (2H, d)

[194] Preparation of Compound G-2

[195] Compound G-1 (60g, 0.20mol), benzamidinehydrochloride (94.2g, 0.60mol), sodium hydroxide (24.1g, 0.60mol), and ethanol (1200mL) were mixed and refluxed for 12 hours. When the reaction was completed, the reactant was cooled to room temperature, and the precipitated solid was filtered under reduced pressure and washed with water and ethanol. The resultant material was dried, thereby obtaining Compound G-2 (43g, 53.7%) as a yellow solid.

[196] ^{1}H NMR ($CDCl_3$) d 8.65 (3H, m), 7.70 (4H, q), 7.51 (5H, m), 7.28 (1H, s), 3.40 (2H, d)

[197] Preparation of Compound G

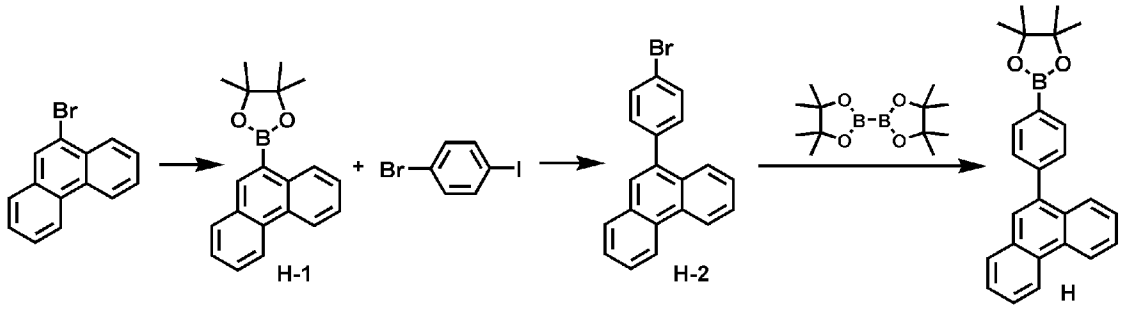
[198] Compound G-2 (50g, 0.13mol), potassium tertiary butoxide (t-BuOK, 29.5g, 0.26mol), and tetrahydrofuran (THF, 750mL) were mixed and stirred at 0°C until the mixture was completely dissolved. Then, methyliodide (MeI, 44.4g, 0.31mol) was slowly added thereto. When the reaction was completed after stirring at room temperature for 4 hours, water is added thereto. Then, the resultant was extracted with MC and dried over anhydrous magnesium sulfate, followed by filtration and concentration under reduce pressure, thereby obtaining Compound G (24g, 44.9%) as a white solid.

[199] ^{1}H NMR ($CDCl_3$) d 8.65 (3H, m), 7.70 (4H, q), 7.51 (5H, m), 7.28 (1H, s), 3.40 (2H, d), 1.67 (6H, s)

[200]

[201] [Preparation Example 8] Preparation of Compound H

[202]



[203] Preparation of Compound H-1

[204] 9-bromophenanthrene (67.2g, 0.26mol), bis(pinacolato)diboron, (69.68g, 0.27mol), 1,4-dioxane (1350mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (3.82g, 5.23mmol), and potassium acetate (51.3g, 0.52mol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over anhydrous magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound H-1 (69.2g, 69.2%) as a solid.

[205] ^{1}H NMR ($CDCl_3$) d 8.89 (d, 1H), 8.86 (d, 1H), 8.11-7.78 (m, 7H) , 1.40 (s, 12H)

[206] Preparation of Compound H-2

[207] Compound H-1 (64.5g, 0.21mol), 1-bromo-4-iodobenzene (60.0g, 0.21mol), $Pd(PPh_3)_4$ (4.9g, 4.2mmol), 2M K_2CO_3 aqueous solution (300mL), and THF (600mL) were mixed and stirred under reflux for 24 hours. After an organic layer was extracted with MC, the extracted organic layer was concentrated under reduced pressure and washed with acetone, thereby obtaining Compound H-2 (60.1g, 84.9%).

[208] ^{1}H NMR ($CDCl_3$) d 8.89 (d, 1H), 8.86 (d, 1H), 8.11-7.78 (m, 11H)

[209] Preparation of the compound H

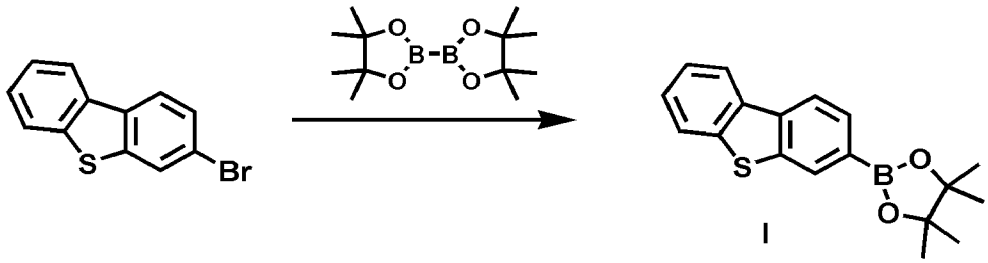
[210] Compound H-2 (60.0g, 0.18mol), bis(pinacolato)diboron, (48.0g, 0.19mol), 1,4-dioxane (1200mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (2.64g, 3.60mmol), and potassium acetate (35.4g, 0.36mol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound H (32.0g, 46.7%) as a solid.

[211] ^{1}H NMR ($CDCl_3$) d 8.89 (d, 1H), 8.86 (d, 1H), 8.11-7.78 (m, 11H), 1.40 (s, 12H)

[212]

[213] [Preparation Example 9] Preparation of Compound I

[214]



I

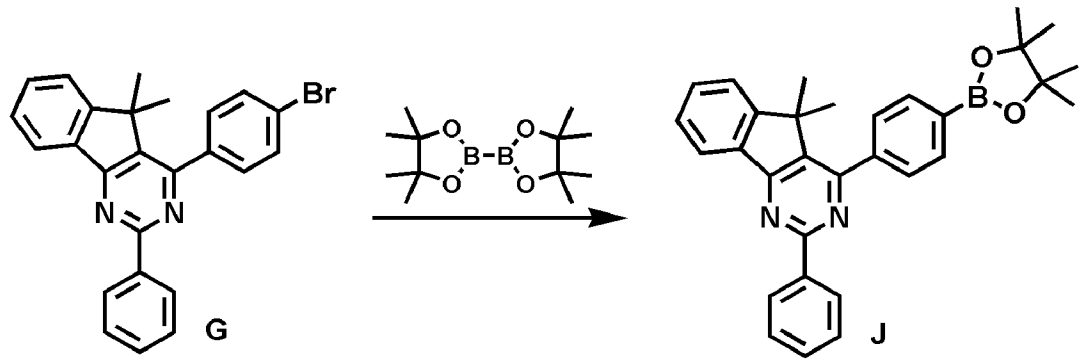
[215] 2-bromodibenzothiophene (50g, 190mmol), bis(pinacolato)diboron, (50.7g, 199.5mmol), 1,4-dioxane (1000mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (2.78g, 3.8mmol), and potassium acetate (37.3g, 380mmol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over anhydrous magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. The filtrate was concentrated under reduced pressure, thereby obtaining Compound I (23g, 39%).

[216] ^{1}H NMR ($CDCl_3$) d 7.88 (m, 4H), 7.32 (m, 3H), 1.40 (s, 12H)

[217]

[218] [Preparation Example 10] Preparation of Compound J

[219]



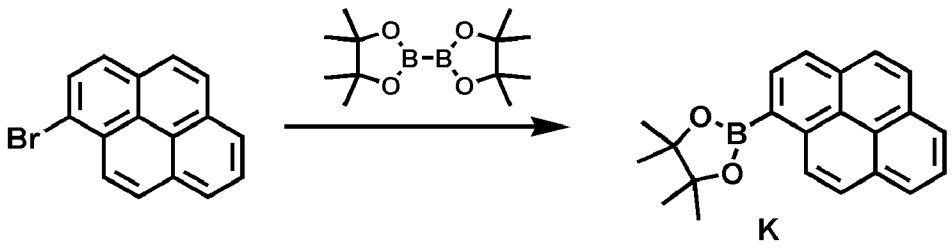
[220] Compound G (10g, 23.4mmol), bis(pinacolato)diboron, (6.5g, 25.5mmol), 1,4-dioxane (200mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (0.2g, 0.3mmol), and potassium acetate (4.6g, 46.8mmol) were mixed and stirred under reflux. After 12 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound J (9.8g, 87.9%) as a yellow solid.

[221] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.68 (4H, q), 7.55 (5H, m), 7.12 (1H, s), 1.40 (s, 12H)

[222]

[223] [Preparation Example 11] Preparation of Compound K

[224]

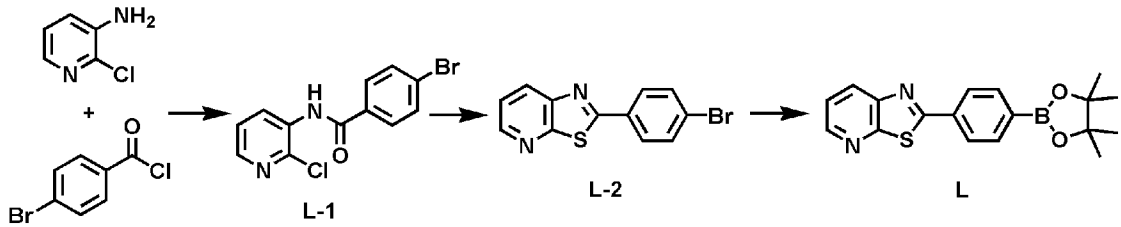


[225] 1-bromopyrene (40g, 142mmol), bis(pinacolato)diboron, (39.74g, 157mmol), 1,4-dioxane (480mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (1.16g, 1.4mmol), and potassium acetate (27.93g, 284mmol) were mixed and stirred under reflux. After 9 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was re-crystallized in ethylacetate, thereby obtaining Compound K (34.09g, 73%).

[226] ^{1}H NMR ($CDCl_3$) d 8.12-8.1 (d, 2H), 7.93-7.70 (m, 5H), 7.71 (d, 2H), 1.26 (q, 12H)

[227]

[228] [Preparation Example 12] Preparation of Compound L

[229] 

[230] Preparation of Compound L-1

[231] Dichloromethane (31mL) and 4-bromobenzoyl chloride (25g, 0.1139mol) were mixed and stirred at -10℃. After 4-bromobenzoyl chloride was completely dissolved, a mixing solution of pyridine (106mL) and 2-chloropyridine-3-amine (13.2g, 0.1026mol) was slowly dropped thereinto and stirred. After 2 hours 30 minutes, water (1250mL) was added thereto, and the prepared solid compound was filtered and washed with methanol. The resultant material was dried, thereby obtaining Compound L-1(28.0g, 87%) as a white solid.

[232] ^{1}H NMR ($CDCl_3$) d 8.43 (d, 1H), 8.00 (s, 1H), 7.84-7.15 (d, 2H), 7.69-7.17 (m, 4H)

[233] Preparation of Compound L-2

[234] 1,3-dimethyl-3,4,5,6-tetrahydro-2(1H)-pyrimidinone (DMPU, 100mL), Compound L-1 (27g, 0.0866mol), and Lawesson s reagent (35.1g) were mixed and stirred under reflux. After 2 hours 30 minutes, an organic layer was extracted with ethylacetate (EA) and dried over magnesium sulfate, followed by filtering. Then, a solid prepared by concentrating the filtrate under reduced pressure was washed with methanol, thereby obtaining Compound L-2 (20g, 80%).

[235] ^{1}H NMR ($CDCl_3$) d 8.59 (d, 1H), 7.75 (d, 1H), 7.49-7.26 (d, 2H), 7.38-7.15 (m, 3H)

[236] Preparation of the compound L

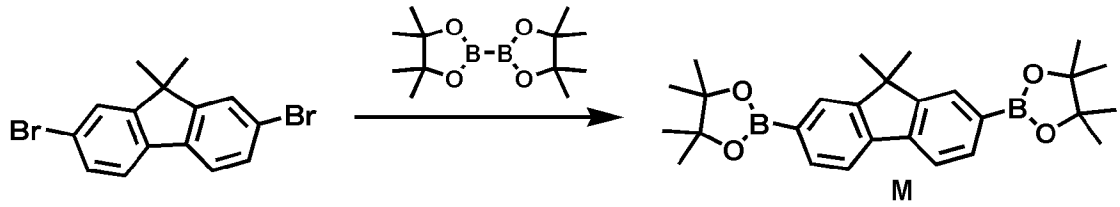
[237] Compound L-2 (27.47g, 103mmol), bis(pinacolato)diboron, (27.47g, 108mmol), 1,4-dioxane (450mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (1.68g, 2.1mmol), and potassium acetate (20.22g, 206.1mmol) were mixed and stirred under reflux for 12 hours. Then, the mixture was cooled to room temperature. An organic layer was extracted by adding saturated sodium chloride (NaCl) aqueous solution and ethylacetate (EA) to the reactant, dried over magnesium sulfate (MgSO4), and treated with activated charcoal, followed by filtering with celite. After a solid prepared by concentrating the filtrate under reduced pressure was suspended and stirred in hexane, the suspension was filtered again and washed with hexane, thereby obtaining Compound L (41.8g, 89 %).

[238] ^{1}H NMR ($CDCl_3$) d 8.59 (d, 1H), 7.75 (d, 1H), 7.50-7.22 (m, 2H), 7.38-7.06 (m, 3H), 1.26 (s, 12H)

[239]

[240] [Preparation Example 13] Preparation of Compound M

[241]



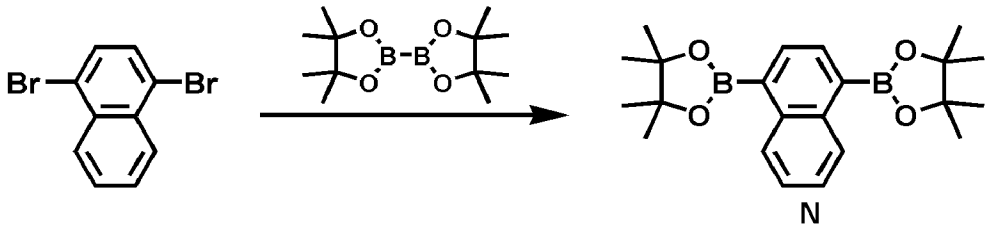
[242] 2.7-dibromo-9,9-dimethyl-9H-fluorene (25g, 71.0mmol), bis(pinacolato)diboron, (37.87g, 149mmol), 1,4-dioxane (300mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (0.58g, 0.7mmol), and potassium acetate (13.94g, 142mmol) were mixed and stirred under reflux. After 9 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was re-crystallized in ethylacetate, thereby obtaining Compound M (30.9g, 61%).

[243] ^{1}H NMR ($CDCl_3$) d 7.80 (m, 2H), 7.60 (m, 2H), 7.40 (m, 2H), 1.67-0.86 (s, 6H), 1.26 (s, 24H)

[244]

[245] [Preparation Example 14] Preparation of Compound N

[246]

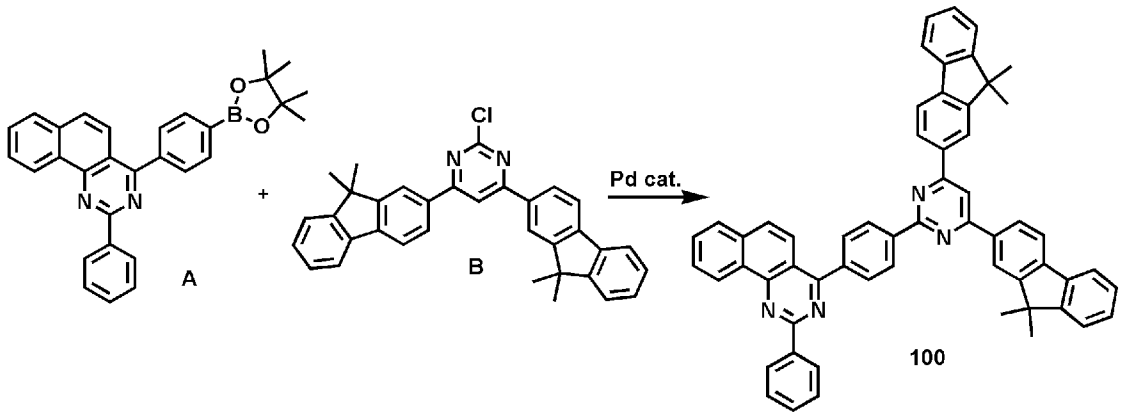


[247] 1,4-dibromonaphthalene (25g, 87.4mmol), bis(pinacolato)diboron, (46.62g, 184mmol), 1,4-dioxane (300mL), 1,1'-bis(diphenylphosphino)ferrocene-palladium(II)dichloride dichloromethane complex (0.71g, 0.9mmol), and potassium acetate (17.16g, 174.8mmol) were mixed and stirred under reflux. After 9 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was re-crystallized in ethylacetate, thereby obtaining Compound N (21.93g, 66%).

[248] ^{1}H NMR ($CDCl_3$) d 7.70-7.67 (m, 2H), 7.30-7.32 (m, 4H), 1.26 (s, 30H)

[249]

[250]



[251] Preparation of Compound 100

[252] Compound A (Preparation Example 1, 9.8g, 23.83mmol), Compound B (Preparation Example 2, 12.0g, 24.1mmol), $Pd(PPh_3)_4$ (0.55g, 0.48mmol), K_2CO_3 (6.59g, 47.65mmol), distilled water (49mL), and THF(98mL) were mixed and stirred under reflux for 12 hours. When the reaction was completed, the reactant was cooled to room temperature, and the prepared solid was washed with water and methanol, thereby obtaining Compound 100 (12.1g, 63.4%).

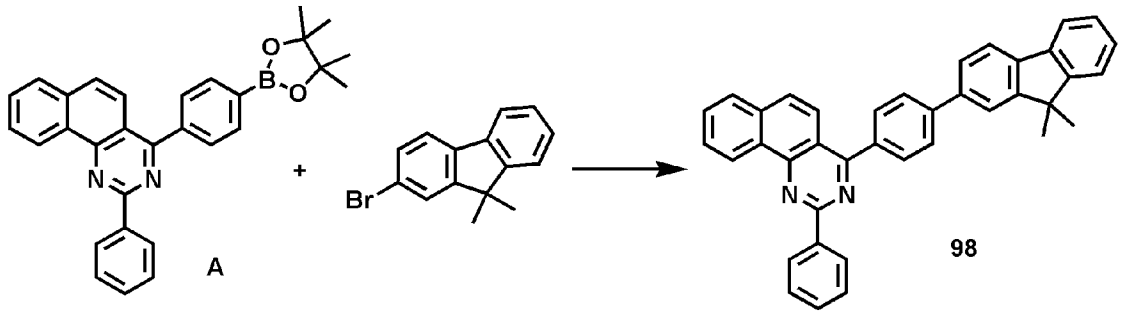
[253] ^{1}H NMR ($CDCl_3$) d 9.61(t, 1H), 9.05(d, 2H), 8.94(d, 2H), 8.47(s, 2H), 8.38(dd, 2H), 8.20(d. 3H), 8.15(d, 1H), 7.98(d, 3H), 7.86(m, 5H), 7.59(m, 5H), 7.44(t, 4H), 1.68(12H)

[254] MALDI-TOF MS: m/z 694.98, cal. 695.61

[255]

[256] [Example 2] Preparation of Compound 98

[257]



[258] 2-bromo-9,9-dimethyl-9H-fluorene (9g, 32.9mmol), Compound A (Preparation Example 1, 15.10g, 32.9mmol), THF (135mL), $Pd(PPh_3)_4$ (0.38g, 0.3mmol), and 1M potassium carbonate aqueous solution (68mL) were mixed and stirred under reflux. After reaction for 18 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, dried over magnesium sulfate, and treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was re-crystallized, thereby obtaining Compound 98 (14.58g, 84%).

[259] ^{1}H NMR ($CDCl_3$) d 8.62(3H,m), 7.78(2H,d), 7.68(6H,q), 7.60(2H,m), 7.55(5H,m),

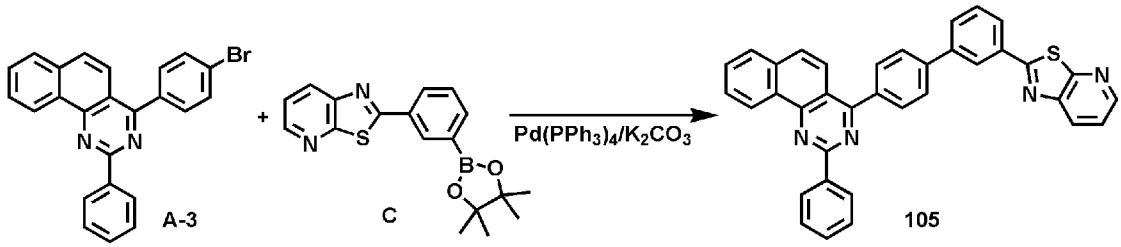
7.42(1H,s), 7.38(3H,m), 1.58 (s, 6H)

[260] MALDI-TOF MS: m/z 524.65, cal. 525.23

[261]

[262] [Example 3] Preparation of Compound 105

[263]



[264] Compound A-3 (Preparation Example 1, 16.87g, 36.8mmol), Compound C (Preparation Example 3, 11.8g, 40.5mmol), THF (300mL), $Pd(PPh_3)_4$ (0.85g, 0.74mmol), and 2M potassium carbonate aqueous solution (55mL) were mixed and stirred under reflux. After reaction for 18 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and ethyl acetate, and dried over magnesium sulfate, followed by concentration under reduced pressure. After the solid obtained after concentration was input in ethyl acetate and suspended under heating, the suspension was filtered and washed with ethyl acetate, thereby obtaining Compound 105 (17.7g, 89%).

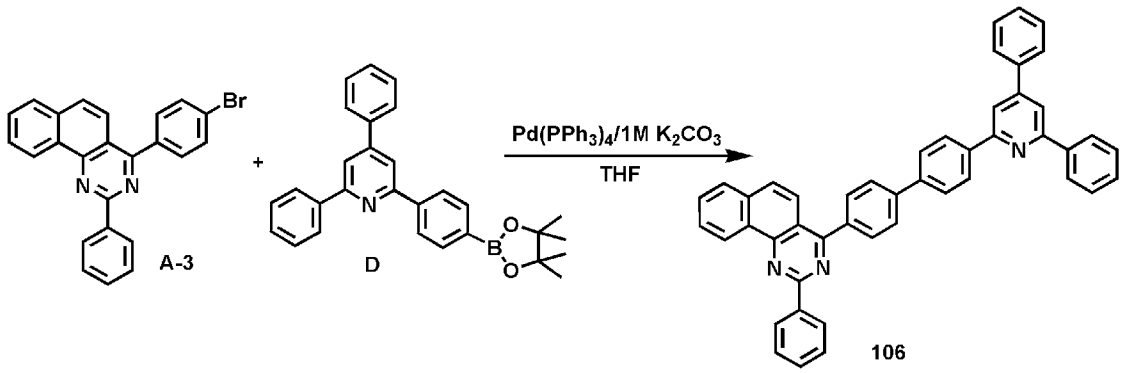
[265] ^{1}H NMR ($CDCl_3$) d 9.62(d, 1H),8.91 (d, 2H), 8.64 (d, 1H), 8.50(s, 1H), 8.38 (d, 1H), 8.16 (d, 1H), 8.11-7.92 (m, 2H), 7.91(d, 2H), 7.88-7.72 (m, 3H), 7.70 (t, 1H), 7.63-7.57 (m, 2H), 7.52 (dd, 1H)

[266] MALDI-TOF MS: m/z 542.652 cal. 542.979

[267]

[268] [Example 4] Preparation of Compound 106

[269]



[270] Compound A-3 (Preparation Example 1, 14g, 34mmol), Compound D (Preparation Example 4, 17.7g, 41mmol), $Pd(PPh_3)_4$ (0.4g, 0.3mmol), THF (150mL), potassium carbonate (27.6g, 199mmol), and water (200mL) were mixed and stirred under reflux. After reaction for 12 hours, the reactant was cooled to room temperature and extracted with methylene chloride, followed by concentration. The resultant material was recrys-

tallized using methanol, thereby obtaining Compound 106 (16.4g, 75%) as a white solid.

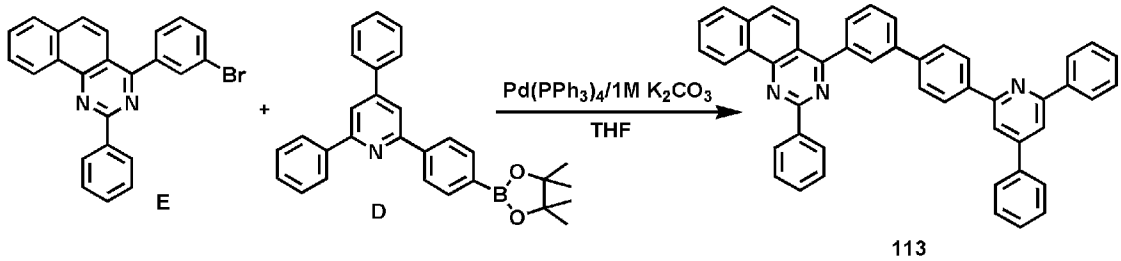
[271] ^{1}H NMR ($CDCl_3$) d 7.50-7.61(m, 9H), 7.80-7.98(m, 12H), 8.08(t, 3H), 8.26(d, 2H), 8.38(d, 2H), 8.89(d, 2H), 9.58(t, 1H)

[272] MALDI-TOF MS: m/z 638.16, cal. 637.77

[273]

[274] [Example 5] Preparation of compound 113

[275]



[276] Compound E (Preparation Example 5, 8g, 19mmol), Compound D (Preparation Example 4, 10.11g, 23mmol), $Pd(PPh_3)_4$ (0.2g, 0.19mmol), THF (150mL), potassium carbonate (13.8g, 99mmol), and water (100mL) were mixed and stirred under reflux. After reaction for 12 hours, the reactant was cooled to room temperature and extracted with methylene chloride, followed by concentration. The resultant material was washed with hexane and ethyl acetate, thereby obtaining Compound 113 (8.6g, 69%) as a light yellow solid.

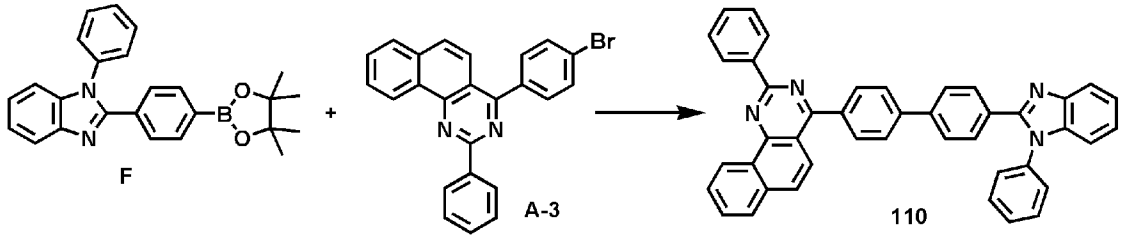
[277] ^{1}H NMR ($CDCl_3$) d 7.44-7.60(m, 9H), 7.73-7.96(m, 14H), 8.06(d, 1H), 8.22(d, 2H), 8.34(d, 2H), 8.88(d, 2H), 9.58(t, 1H)

[278] MALDI-TOF MS: m/z 638.07, cal. 637.77

[279]

[280] [Example 6] Preparation of Compound 110

[281]



[282] Compound F (Preparation Example 6, 14.74g, 37.2mmol), Compound A-3 (Preparation Example 1, 15.3g, 37.2mmol), THF (230mL), $Pd(PPh_3)_4$ (0.43g, 0.4mmol), and 1M potassium carbonate aqueous solution (115mL) were mixed and stirred under reflux. After reaction for 18 hours, the reactant was cooled to room temperature, and an organic layer was extracted by adding saturated sodium chloride aqueous solution and dichloromethane, dried over magnesium sulfate, and treated with activated charcoal, followed by filtering with celite. A solid obtained by concentrating

the filtrate under reduced pressure was re-crystallized, thereby obtaining Compound 110 (15.47g, 69%).

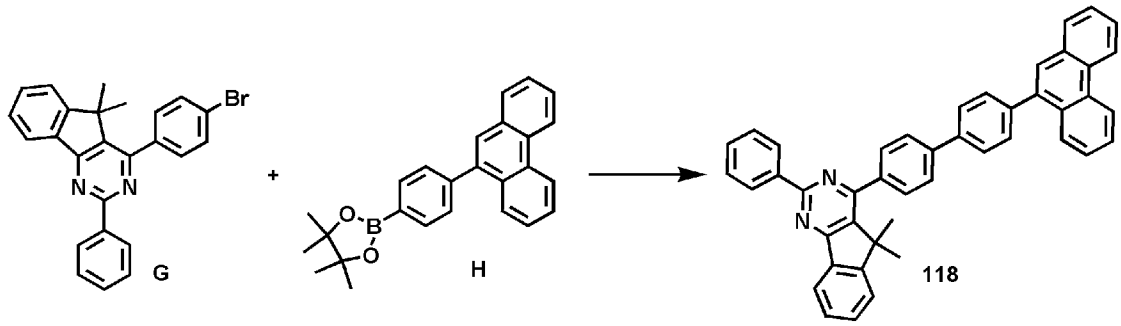
[283] ^{1}H NMR ($CDCl_3$) d 8.81(d, 2H), 8.64(d,2H), 7.92(d, 1H), 7.75(d, 2H), 7.61(d, 5H), 7.59-7.38(m, 7H), 7.36-7.24(m, 7H), 7.19(s, 2H)

[284] MALDI-TOF MS: m/z 601.32 , cal. 600.71

[285]

[286] [Example 7] Preparation of Compound 118

[287]



[288] Compound G (Preparation Example 7, 10g, 23.4mmol), Compound H (Preparation Example 8, 8.9g, 23.4mmol), THF (150mL), $Pd(PPh_3)_4$ (0.54g, 0.47mmol), K_2CO_3 (19.4g, 140.4mmol), and distilled water (75mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered and washed with methanol. The prepared white solid was filtered with celite in toluene with heating. A solid prepared by concentrating the filtrate under reduced pressured was washed with methanol and suspended, followed by filtering, thereby obtaining Compound 118 (11.5g, 81.8%).

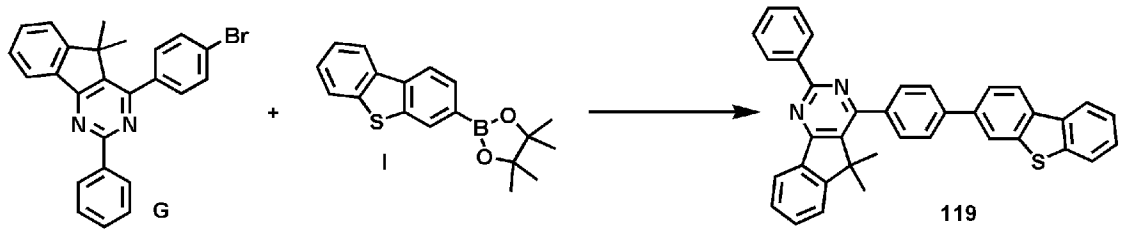
[289] ^{1}H NMR ($CDCl_3$) d 8.89 (d, 1H), 8.86 (d, 1H), 8.65 (3H, m), 8.11-7.78 (m, 11H), 7.70 (4H, q), 7.51 (5H, m), 7.28 (1H, s), 3.40 (2H, d)

[290] MALDI-TOF MS: m/z 600.71, cal. 600.75

[291]

[292] [Example 8] Preparation of Compound 119

[293]



[294] Compound G (Preparation Example 7, 10g, 23.4mmol), Compound I (Preparation Example 9, 8.9g, 23.4mmol), THF (150mL), $Pd(PPh_3)_4$ (0.54g, 0.47mmol), K_2CO_3 (19.4g, 140.4mmol), and distilled water (75mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered and washed with methanol. The

prepared white solid was filtered with celite in toluene with heating. A solid prepared by concentrating the filtrate under reduced pressured was washed with methanol and suspended, followed by filtering, thereby obtaining Compound 119 (13g, 91%).

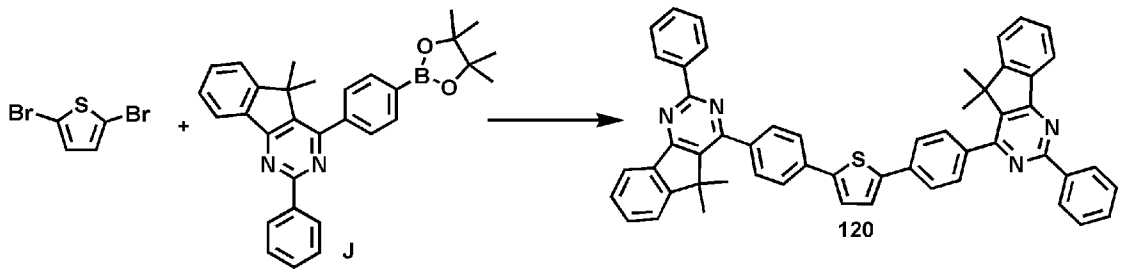
[295] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.88 (m, 4H), 7.68 (4H, q), 7.55 (5H, m), 7.32 (m, 3H), 7.12 (1H, s), 1.40 (s, 12H)

[296] MALDI-TOF MS: m/z 310.18, cal. 310.22

[297]

[298] [Example 9] Preparation of Compound 120

[299]



[300] 1,3-dibromothiophene (10g, 41.3mmol), Compound J (Preparation Example 10, 39.2g, 82.7mmol), THF (200mL), $Pd(PPh_3)_4$ (1.91g, 1.65mmol), K_2CO_3 (68.6g, 496mmol), and distilled water (100mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered and washed with methanol, thereby obtaining Compound 120 (22g, 68.5%).

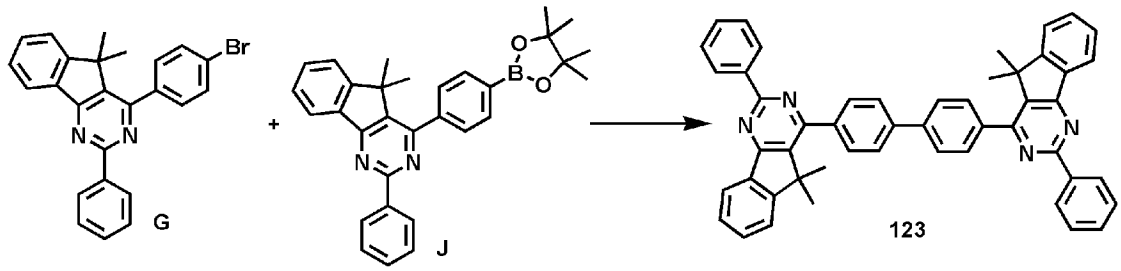
[301] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.68 (4H, q), 7.55 (5H, m), 7.12 (1H, s), 6.64 (d, 1H)

[302] MALDI-TOF MS: m/z 776.95, cal. 776.99

[303]

[304] [Example 10] Preparation of Compound 123

[305]



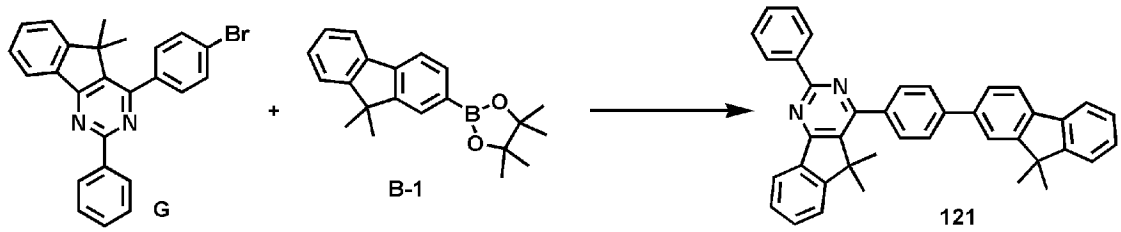
[306] Compound G (Preparation Example 7, 15g, 35.1mmol), Compound J (Preparation Example 10, 16.65g, 35.1mmol), THF (250mL), $Pd(PPh_3)_4$ (0.81g, 0.70mmol), K_2CO_3 (29.11g, 210.6mmol), and distilled water (125mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered and washed with methanol, thereby obtaining Compound 123 (21.7g, 88.97%).

[307] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.68 (4H, q), 7.55 (5H, m), 7.12 (1H, s)

[308] MALDI-TOF MS: m/z 694.84, cal. 694.86

[309]

[310] [Example 11] Preparation of Compound 121

[311] 

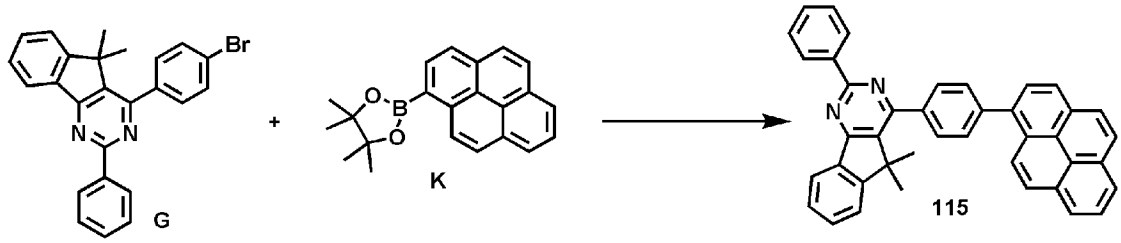
[312] Compound G (Preparation Example 7, 13g, 30.4mmol), Compound B-1 (Preparation Example 2, 9.7g, 30.4mmol), THF (200mL), $Pd(PPh_3)_4$ (0.7g, 0.61mmol), K_2CO_3 (25.2g, 182.5mmol), and distilled water (100mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, the precipitated solid was filtered and washed with methanol, thereby obtaining Compound 121 (14.2g, 86.33%).

[313] ^{1}H NMR ($CDCl_3$) d 8.62 (3H, m), 7.78 (2H, d), 7.68 (4H, q), 7.55 (7H, m), 7.38 (3H, m), 7.12 (1H, s)

[314] MALDI-TOF MS: m/z 540.58, cal. 540.70

[315]

[316] [Example 12] Preparation of Compound 115

[317] 

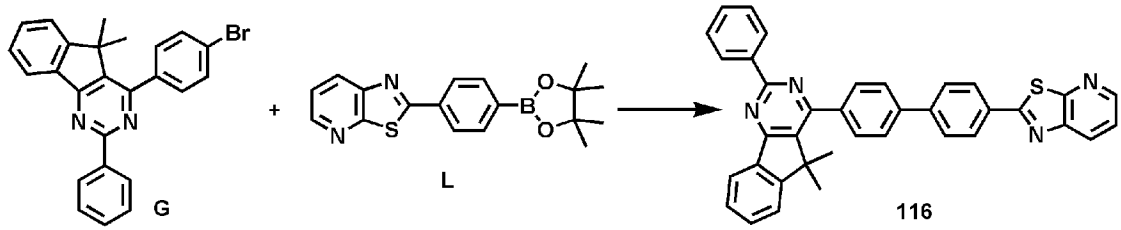
Compound G (Preparation Example 7, 24g, 56.2mmol), Compound K (Preparation Example 11, 18.43g, 56.2mmol), THF (360mL), $Pd(PPh_3)_4$ (0.65g, 0.56mmol), and 1M K_2CO_3 aqueous solution (180mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, an organic layer was extracted with saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was suspended in ethyl acetate with heating, followed by filtering, thereby obtaining Compound 115 (21.8g, 71%).

[318] ^{1}H NMR ($CDCl_3$) d 8.18-8.12 (d, 2H), 8.04-7.82 (d, 1H), 7.88-7.80 (d, 2H), 7.71-7.65 (m, 4H), 7.54-7.26 (d, 4H), 7.48-7.16 (d, 2H), 7.40-7.22 (d, 1H), 7.32-7.26 (m, 3H), 7.19-7.13 (d, 1H), 7.14-7.18 (d, 2H), 1.67 (s, 6H)

[319] MALDI-TOF MS:m/z 548.7, cal. 548.67

[320]

[321] [Example 13] Preparation of Compound 116



[322] Compound G (Preparation Example 7, 24g, 56.2mmol), Compound L (Preparation Example 12, 19g, 56.2mmol), THF (360mL), $Pd(PPh_3)_4$ (0.65g, 0.56mmol), and 1M K_2CO_3 aqueous solution (180mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, an organic layer was extracted with saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was suspended in ethyl acetate with heating, followed by filtering, thereby obtaining Compound 116 (22.9g, 73%).

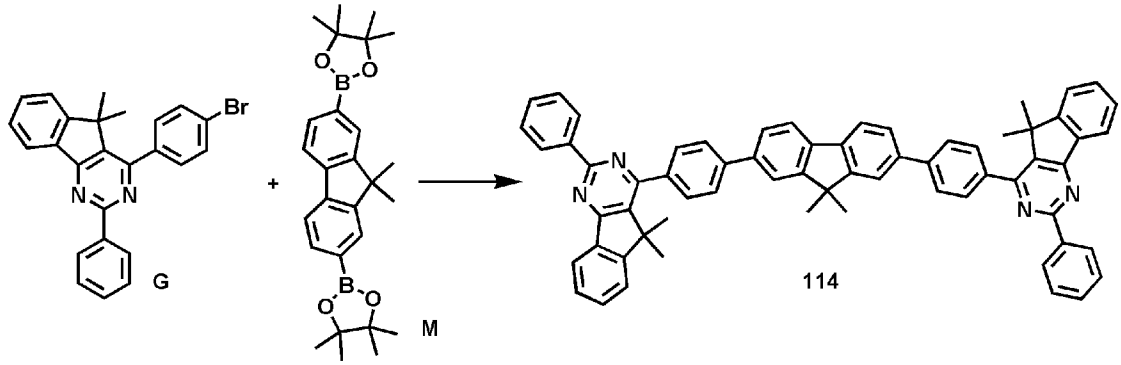
[323] ^1H NMR ($CDCl_3$) d 8.59 (d, 1H), 7.75 (d, 1H), 7.54-7.26 (m, 10H), 7.40-7.22 (m, 5H), 7.19-7.06 (m, 3H), 1.67 (s, 6H)

[324] MALDI-TOF MS : m/z 558.8, cal. 558.69

[325]

[326] [Example 14] Preparation of Compound 114

[327]



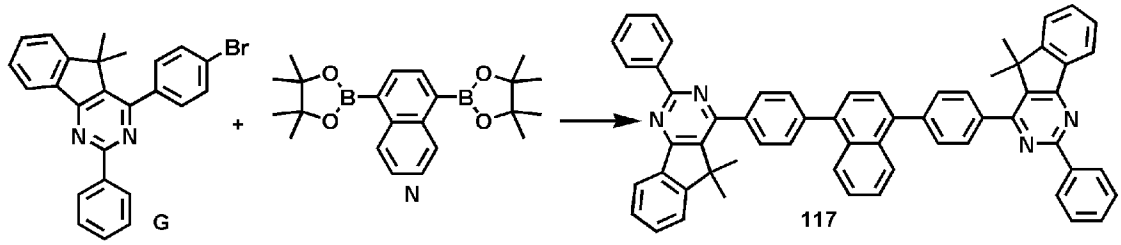
[328] Compound G (Preparation Example 7, 24g, 56.2mmol), Compound M (Preparation Example 13, 15g, 33.6mmol), THF (225mL), $Pd(PPh_3)_4$ (0.39g, 0.336mmol), and 1M K_2CO_3 aqueous solution (113mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, an organic layer was extracted with saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and then treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was suspended in ethyl acetate with heating, followed by filtering, thereby obtaining Compound 114 (21.56g, 70%).

[329] ^{1}H NMR ($CDCl_3$) d 7.90-7.84 (d, 2H), 7.77-7.38 (d, 4H), 7.54-7.26 (m, 12H), 7.40-7.22 (d, 2H), 7.32-7.06 (m, 6H), 7.19-7.04 (m, 6H), 1.67 (s, 24H)

[330] LC-MS:m/z 887.2, cal. 887.12

[331]

[332] [Example 15] Preparation of Compound 117

[333] 

[334] Compound G (Preparation Example 7, 35.42g, 82.9mmol), Compound N (Preparation Example 14, 15g, 39.5mmol), THF (225mL), $Pd(PPh_3)_4$ (0.46g, 0.395mmol), and 1M K_2CO_3 aqueous solution (113mL) were mixed and stirred under reflux for 18 hours. When the reaction was completed, the reactant was cooled to room temperature. Then, an organic layer was extracted with saturated sodium chloride aqueous solution and ethylacetate, dried over magnesium sulfate, and treated with activated charcoal, followed by filtering with celite. A solid prepared by concentrating the filtrate under reduced pressure was suspended in ethyl acetate with heating, followed by filtering, thereby obtaining Compound 117 (19.4g, 60%).

[335] ^{1}H NMR ($CDCl_3$) d 7.67 (d, 2H), 7.60-7.32 (d, 2H), 7.54-7.26 (m, 8H), 7.48-7.22 (m, 6H), 7.32-7.06 (m, 6H), 7.22-7.04 (m, 4H), 7.14-7.01 (m, 4H), 1.67 (s, 12H)

[336] MALDI-TOF MS:m/z 821.1, cal. 821.02

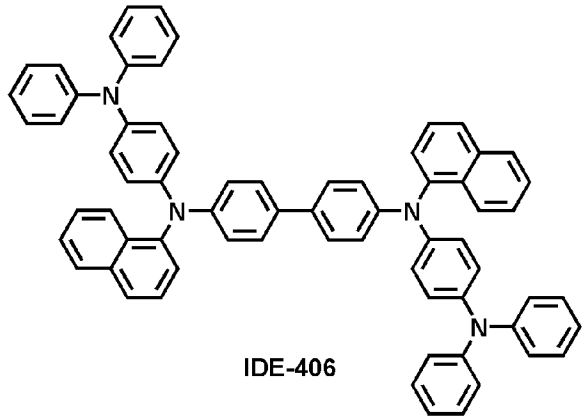
[337]

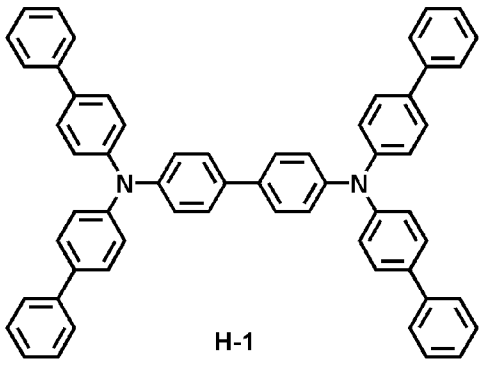
[338] [Example 16] Manufacturing organic electroluminescent device using Compound 100 according to the present invention

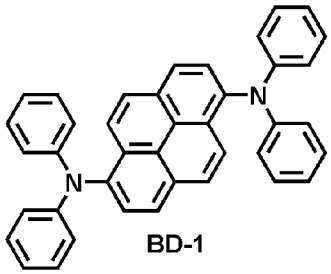
[339] A glass substrate (25mm x 25mm x 0.7mm) having an indium tin oxide (ITO) transparent electrode line having a thickness of 150nm was subjected to ultrasonic cleaning for 10 minutes in distilled water in which a detergent was dissolved and then cleaned with distilled water again for 10 minutes. After washing with distilled water, the substrate was subjected to ultrasonic cleaning with solvents in a sequence of isopropyl alcohol, acetone, and methanol for 10 minutes, respectively, and dried. Thereafter, the substrate was dry cleaned using oxygen/argon plasma, then the glass substrate having the transparent electrode line was mounted on a substrate holder of a vacuum vapor deposition apparatus. A film having a thickness of 60nm was formed on a surface on which the transparent electrode line was formed as a hole injection layer using IDE-406 (the following structure, Idemitsu) so as to cover the transparent electrode. Next, a film having a thickness of 30nm was formed on the IDE-406 film as

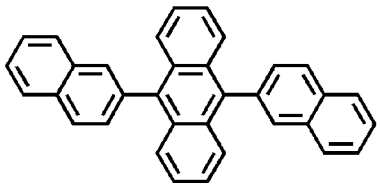
a hole transport layer using H-1 (tetrakis-N-biphenyl-4-yl-benzidine, hereinafter, referred to as the H-1 film). Then, BD-1 having the following structure and β-ADN(9,10-di(naphthalene-2-yl)anthracene) were deposited on the H-1 film as a dopant and a luminescent host at a weight ratio of 5%, to thereby form a film having a thickness of 20nm as a luminescent layer.

[340]









[341] A film having a thickness of 20nm was formed on the luminescent layer as an electron transport layer by depositing Compound 100 according to the present invention. Subsequently, lithium quinolate (Liq) was deposited thereon to form an electron injection layer. Metal aluminum was deposited on this Liq film to form a metal cathode, thereby manufacturing the organic electroluminescent device.

[342]

[343] [Example 17] Manufacturing organic electroluminescent device using Compound 98 according to the present invention

[344] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 98 was used as an electron transport material instead of Compound 100 in Example 16.

[345]

[346] [Example 18] Manufacturing organic electroluminescent device using Compound 105 according to the present invention

[347] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 105 was used as an electron transport material instead of Compound 100 in Example 16.

[348]

[349] [Example 19] Manufacturing organic electroluminescent device using Compound 106 according to the present invention

[350] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 106 was used as an electron transport material instead of Compound 100 in Example 16.

[351]

[352] [Example 20] Manufacturing organic electroluminescent device using Compound 113 according to the present invention

[353] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 113 was used as an electron transport material instead of Compound 100 in Example 16.

[354]

[355] [Example 21] Manufacturing organic electroluminescent device using Compound 110 according to the present invention

[356] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 110 was used as an electron transport material instead of Compound 100 in Example 16.

[357]

[358] [Example 22] Manufacturing organic electroluminescent device using Compound 114 according to the present invention

[359] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 114 was used as an electron transport material instead of Compound 100 in Example 16.

[360]

[361] [Example 23] Manufacturing organic electroluminescent device using Compound 115 according to the present invention

[362] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 115 was used as an electron transport material instead of Compound 100 in Example 16.

[363]

[364] [Example 24] Manufacturing organic electroluminescent device using Compound 116 according to the present invention

[365] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 116 was used as an electron transport material instead of Compound 100 in Example 16.

[366]

[367] [Example 25] Manufacturing organic electroluminescent device using Compound 117 according to the present invention

[368] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 117 was used as an electron transport material instead of Compound 100 in Example 16.

[369]

[370] [Example 26] Manufacturing organic electroluminescent device using Compound 118 according to the present invention

[371] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 118 was used as an electron transport material instead of Compound 100 in Example 16.

[372]

[373] [Example 27] Manufacturing organic electroluminescent device using Compound 119 according to the present invention

[374] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 119 was used as an electron transport material instead of Compound 100 in Example 16.

[375]

[376] [Example 28] Manufacturing organic electroluminescent device using Compound 120 according to the present invention

[377] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 120 was used as an electron transport material instead of Compound 100 in Example 16.

[378]

[379] [Example 29] Manufacturing organic electroluminescent device using Compound 121 according to the present invention

[380] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 121 was used as an electron transport material instead of Compound 100 in Example 16.

[381]

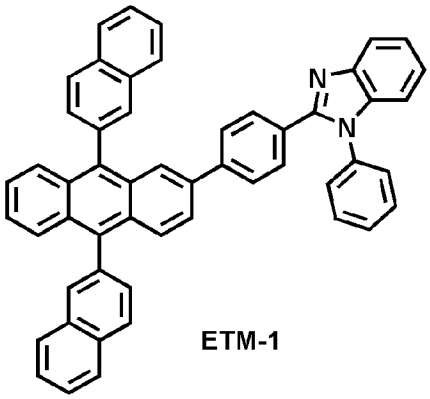
[382] [Example 30] Manufacturing organic electroluminescent device using Compound 123 according to the present invention

[383] An organic electroluminescent device was manufactured by the same process in Example 16 except that Compound 123 was used as an electron transport material instead of Compound 100 in Example 16.

[384] [Comparative Example 1] Manufacturing organic electroluminescent device using Compound ETM-1

[385] An organic electroluminescent device was manufactured by the same process in Example 7 except that Compound ETM-1 having the following structure was used as an electron transport material instead of Compound 100 in Example 7.

[386]



ETM-1

[387]

[388] Measurement results of electroluminescence properties and basic physical properties of the organic electroluminescent devices manufactured in Examples 16 to 30 and Comparative Example 1 were shown in Table 1, and a graph of efficiency (cd/A) and luminance (cd/m^2) of the organic electroluminescent devices manufactured in Examples 16 to 21 and Comparative Example 1 was shown in FIG. 1. In addition, a graph of efficiency (cd/A) and luminance (cd/m^2) of the organic electroluminescent devices manufactured in Examples 22 to 30 and Comparative Example 1 was shown in FIG. 2.

[389]

[390] Table 1

[Table 1]

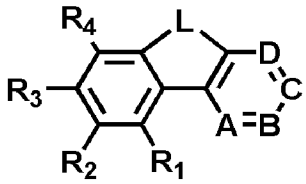
No.	Voltage (V)	Current density (mA/cm^2)	Efficiency (cd/A)	Color coordinate (x,y)	Luminance (cd/m^2)
Example 16	4.60	14.71	7.46	(0.137, 0.151)	1099
Example 17	4.80	14.14	8.06	(0.136, 0.153)	1140
Example 18	4.80	15.07	6.86	(0.138, 0.156)	1035
Example 19	4.80	15.64	6.54	(0.136, 0.149)	1024
Example 20	4.80	13.65	7.35	(0.135, 0.151)	1004
Example 21	5.20	20.64	6.07	(0.135, 0.148)	1255
Example 22	4.8	22.66	4.5	(0.13, 0.14)	1040
Example 23	4.8	16.58	6.8	(0.13, 0.14)	1130
Example 24	7.2	16.17	6.5	(0.13, 0.17)	1067
Example 25	6.4	19.93	5.4	(0.13, 0.14)	1081
Example 26	4.4	28.93	3.4	(0.13, 0.14)	1002
Example 27	7.2	17.87	6.0	(0.13, 0.14)	1077
Example 28	4.8	13.42	7.5	(0.13, 0.17)	1011
Example 29	4.8	15.07	6.8	(0.13, 0.15)	1035
Example 30	5.2	22.28	5.6	(0.13, 0.14)	1267
Comparative Example 1	5.00	29.05	4.03	(0.135, 0.143)	1170

[391] As shown in Table 1, it may be confirmed that the material according to the present invention had excellent luminescence properties as compared with the material according to the related art. In addition, the organic electroluminescent device using the heteroaromatic cyclic compound according to the present invention as the electron transport layer had an excellent luminescence property and decreased driving current to increase the power efficiency, thereby using less consumption power.

Claims

[Claim 1] An electron transport material represented by the following Chemical Formula 1.

[Chemical Formula 1]



[In Chemical Formula 1,

A, B, C, and D each are independently C-$(L_1)_m$-Ar_1 or N, but two of A, B, C, and D are N, the other two are C-$(L_1)_m$-Ar_1, and each of -$(L_1)_m$-Ar_1 are the same as or different from each other, but two -$(L_1)_m$-Ar_1 are not hydrogen at the same time;

L is (C1-C20)alkylene or (C2-C20)alkenylene, a carbon atom (-CH_2-) of alkylene of L is substituted with a heteroatom selected from NR(R is (C1-C30)alkyl), O, and S, and a carbon atom (=CH-) of alkenylene of L is substituted with N;

R_1 to R_4 each are independently hydrogen, (C1-C30)alkyl, (C3-C30)cycloalkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

L_1(s) each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

m is an integer of 1 to 3, and when m is an integer of 2 or more, L_1(s) are the same as or different from each other;

Ar_1(s) each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

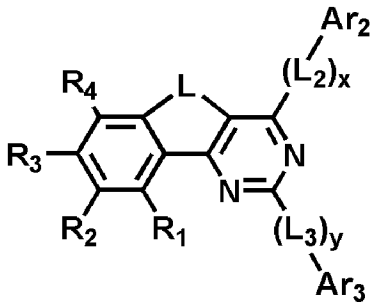
alkyl, cycloalkyl, aryl, heteroaryl of R_1 to R_4, alkylene or alkenylene of L, arylene and heteroarylene of L_1, and aryl and heteroaryl of Ar_1 are substituted with at least one selected from a group consisting of (C1-C30)alkyl, halo(C1-C30)alkyl, halogen, cyano, (C3-C30)cycloalkyl, (C1-C30)alkoxy, (C6-C30)aryloxy, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, (C3-C30)heteroaryl, (C3-C30)heteroaryl substituted with (C1-C30)alkyl, (C3-C30)heteroaryl substituted with (C6-C30)aryl, mono or di(C1-C30)alkylamino, mono or di(C6-C30)arylamino, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, nitro, and hydroxy; and

heteroarylene and heteroaryl contains at least one hetero atom selected

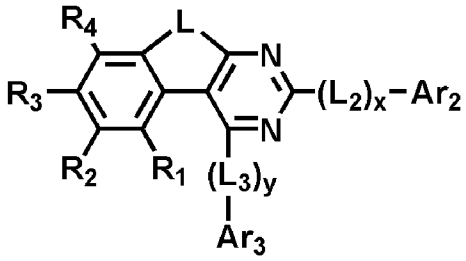
from B, N, O, S, P(=O), Si, and P.

[Claim 2] The electron transport material of claim 1, wherein it is represented by the following Chemical Formula 2 or 3.

[Chemical Formula 2]



[Chemical Formula 3]



[In Chemical Formulas 2 and 3,

R_1 to R_4 and L each has the same definition in Chemical Formula 1 of claim 1;

L_2 and L_3 each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

x and y each are independently an integer of 1 to 3, wherein when x is an integer of 2 or more, L_2(s) are the same as or different from each other, and when y is an integer of 2 or more, L_3(s) are the same as or different from each other;

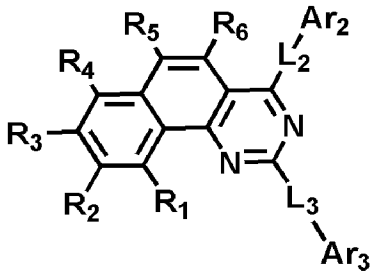
Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

arylene and heteroarylene of L_2 and L_3, and aryl and heteroaryl of Ar_2 and Ar_3 are substituted with at least one selected from a group consisting of (C1-C30)alkyl, halo(C1-C30)alkyl, halogen, cyano, (C3-C30)cycloalkyl, (C1-C30)alkoxy, (C6-C30)aryloxy, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, (C3-C30)heteroaryl, (C3-C30)heteroaryl substituted with (C1-C30)alkyl, (C3-C30)heteroaryl substituted with (C6-C30)aryl, mono or di(C1-C30)alkylamino, mono or di(C6-C30)arylamino, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl,

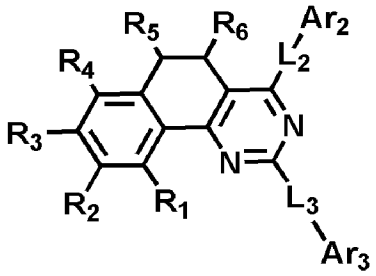
tri(C6-C30)arylsilyl, nitro, and hydroxy; and -$(L_2)_x$-Ar_2 and -$(L_3)_Y$-Ar_3 are not hydrogen at the same time.]

[Claim 3] The electron transport material of claim 2, wherein it is represented by the following Chemical Formulas 4 to 9.

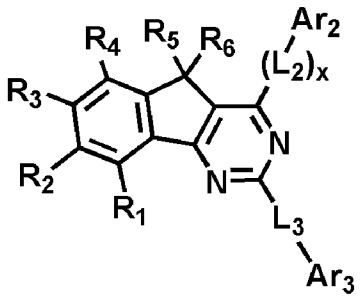
[Chemical Formula 4]



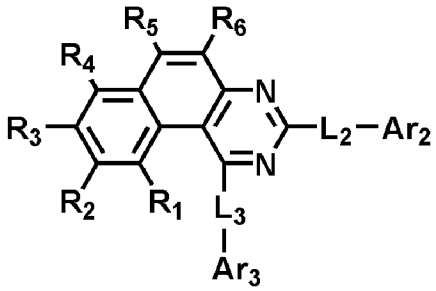
[Chemical Formula 5]



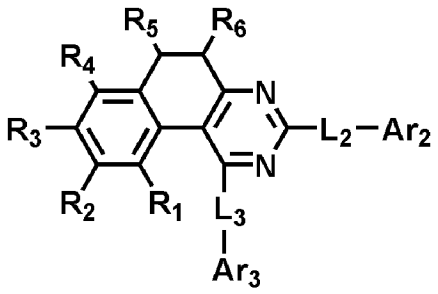
[Chemical Formula 6]



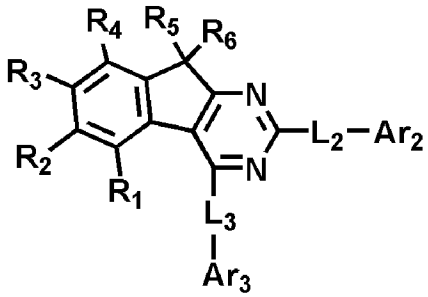
[Chemical Formula 7]



[Chemical Formula 8]



[Chemical Formula 9]



[In Chemical Formulas 4 to 9,

R_1 to R_6 each are independently hydrogen, (C1-C30)alkyl, (C3-C30)cycloalkyl, (C6-C30)aryl, or (C3-C30)heteroaryl;

L_2 and L_3 each are independently a single bond, (C6-C30)arylene, or (C3-C30)heteroarylene;

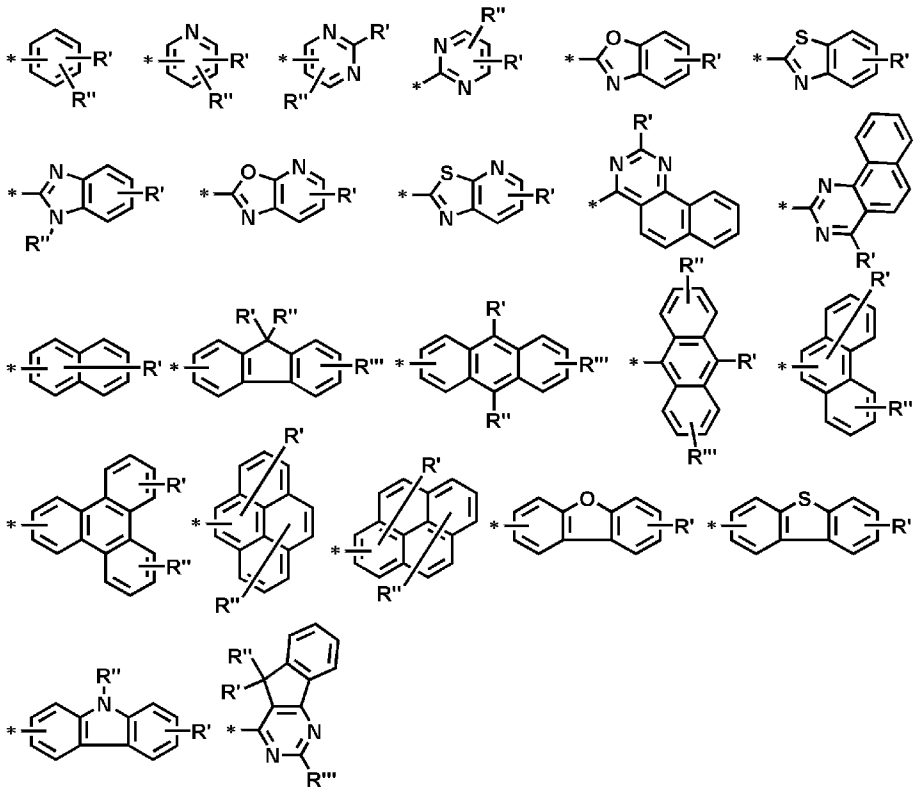
x is an integer of 1 to 3, and when x is an integer of 2 or more, L_2(s) are the same as or different from each other;

Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, or (C3-C30)heteroaryl; and

arylene of L_2 and L_3 and aryl and heteroaryl of Ar_2 and Ar_3 are further substituted with at least one selected from a group consisting of (C1-C30)alkyl, (C6-C30)aryl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, and (C3-C30)heteroaryl.]

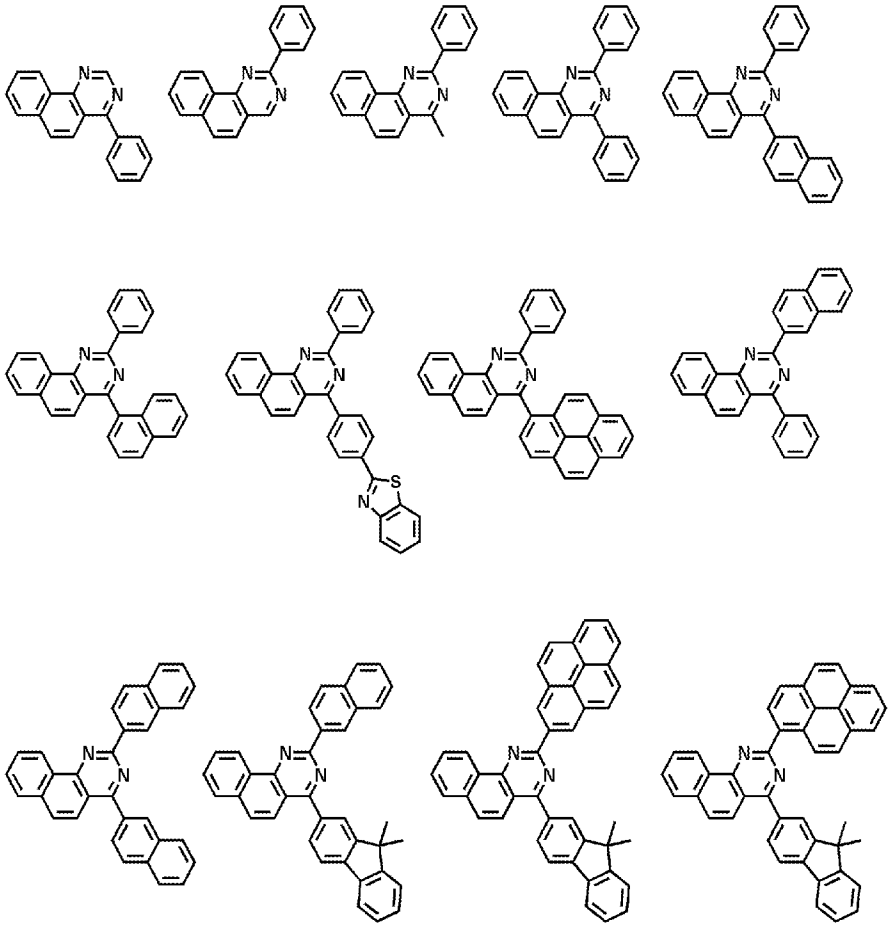
[Claim 4] The electron transport material of claim 3, wherein L_2 and L_3 each are a single bond, phenylene, biphenylene, 9,9-dimethylfluorenylene, naphthylene, anthrylene, pyridinylene, or pyrimidinylene;

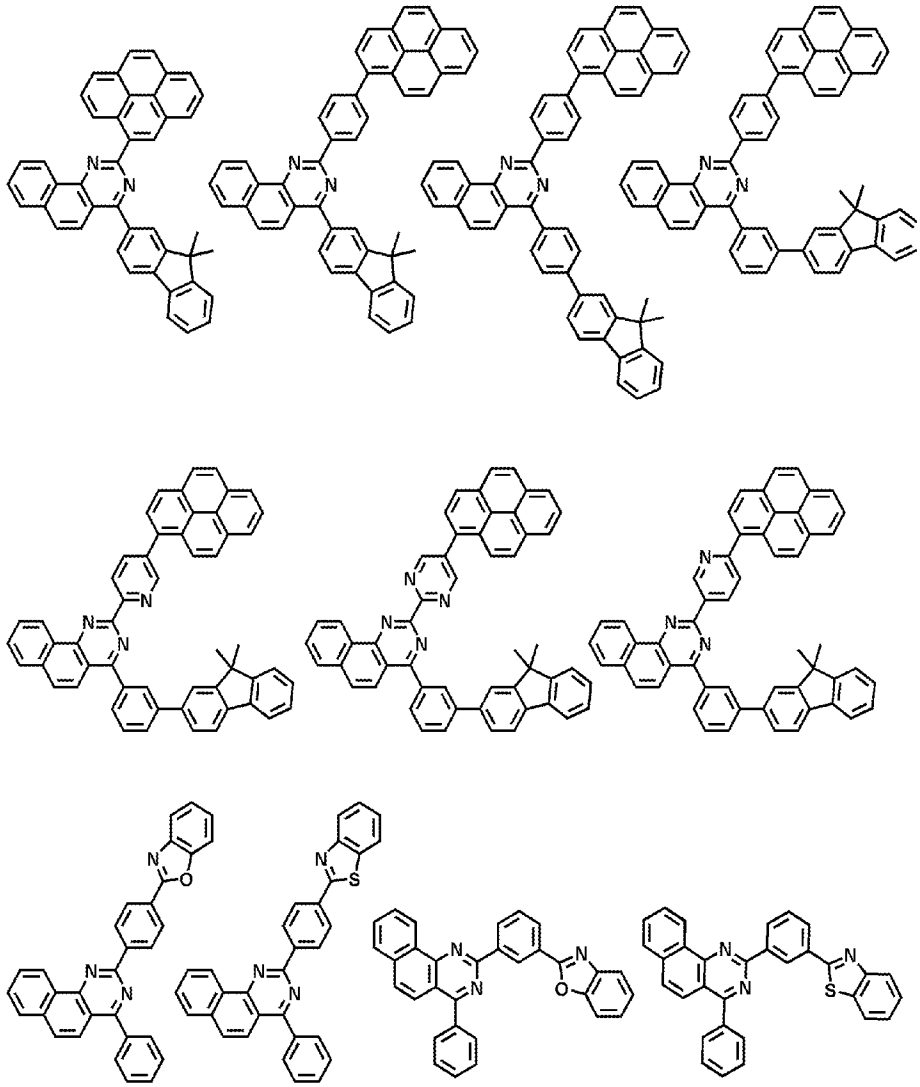
Ar_2 and Ar_3 each are independently hydrogen, (C1-C30)alkyl, or selected from the following structure; and

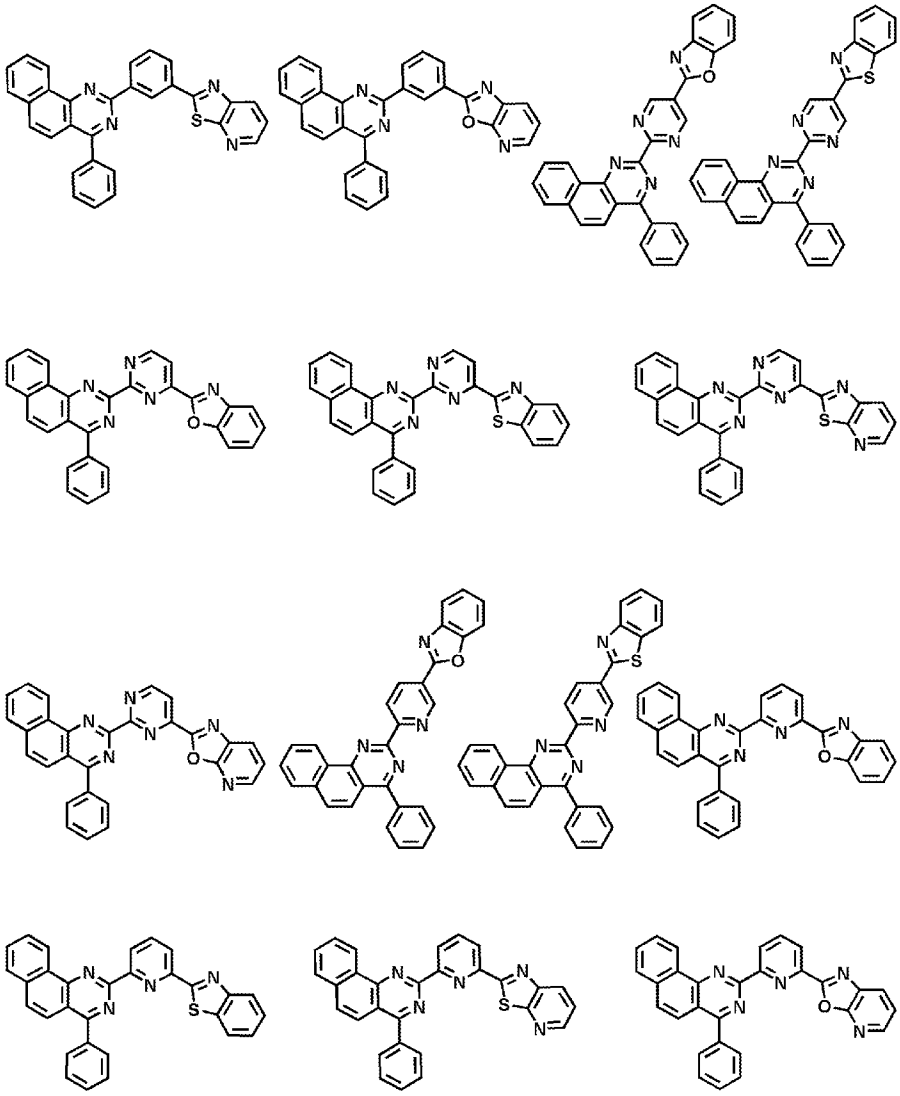


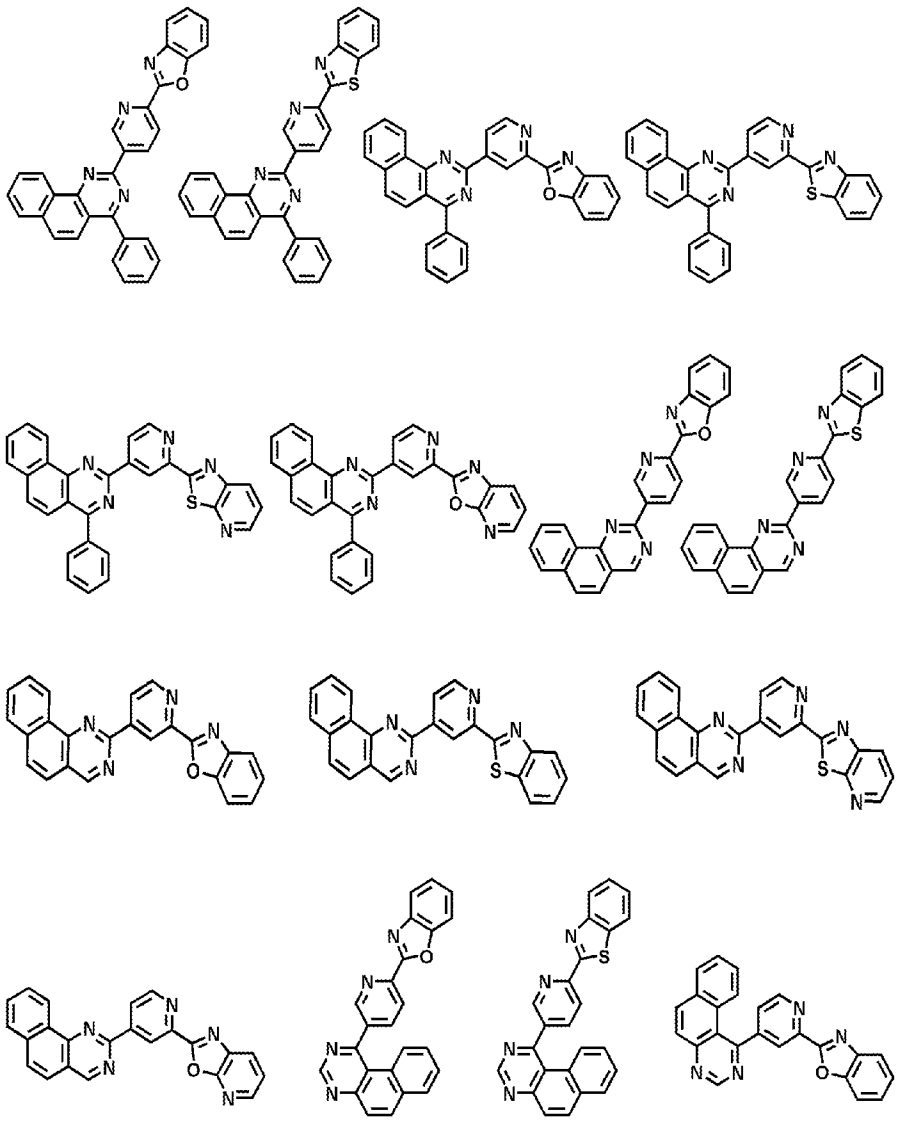
R', R'', and R''' each are independently hydrogen, (C1-C30)alkyl, (C6-C30)aryl, (C3-C30)heteroaryl, or (C1-C30)alkyl(C6-C30)aryl.

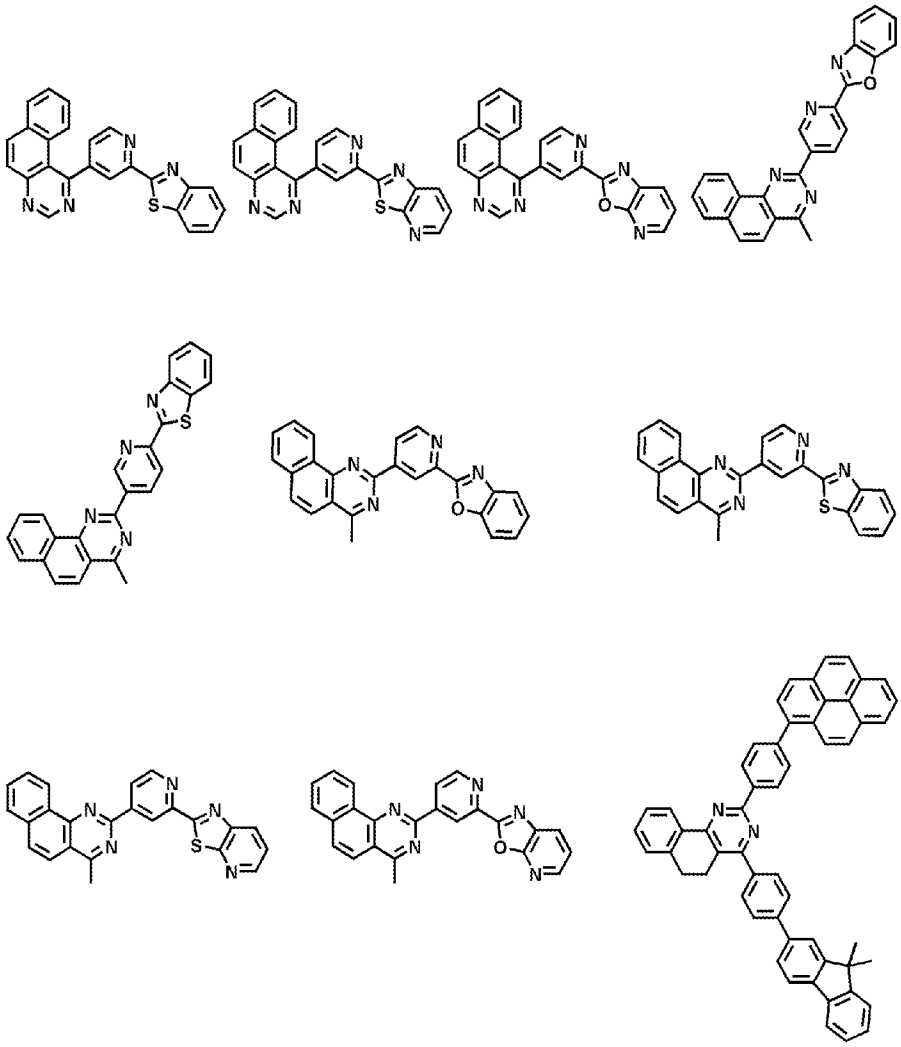
[Claim 5] The electron transport material of claim 4, wherein it is selected from the following compounds.

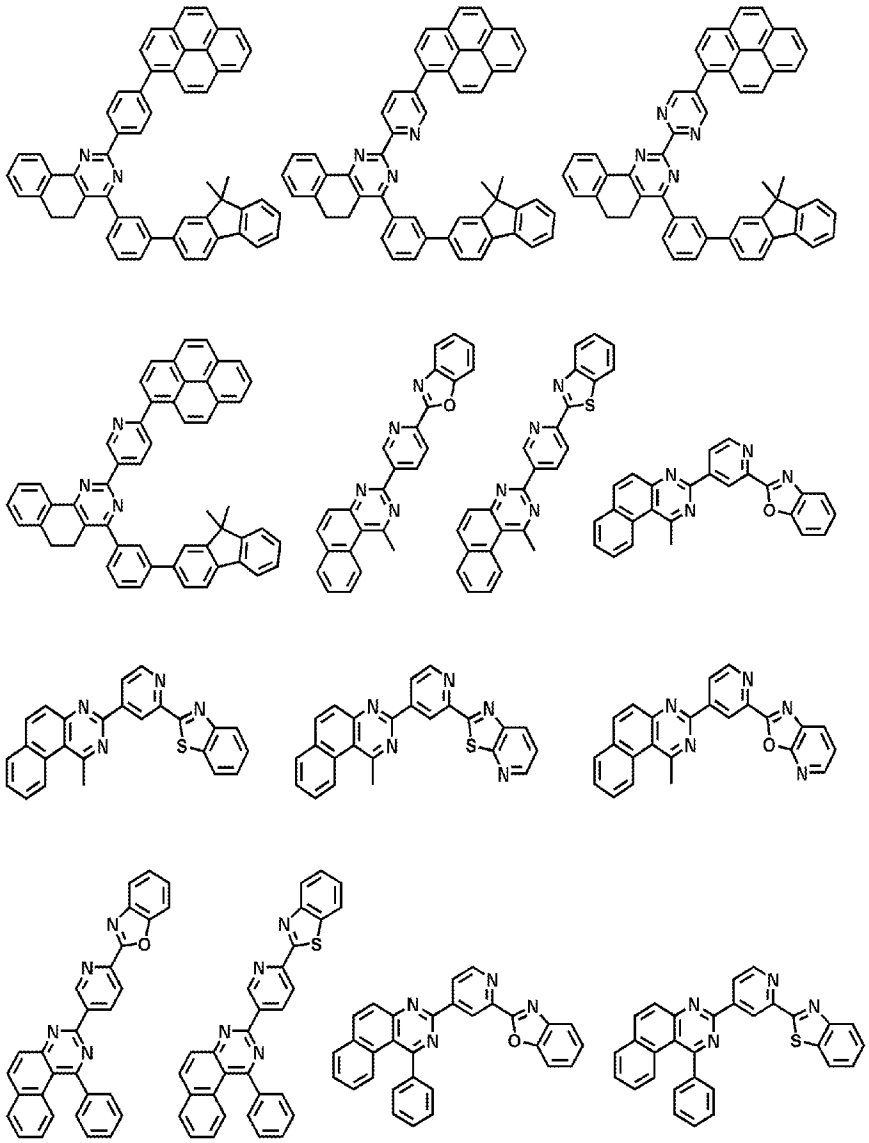


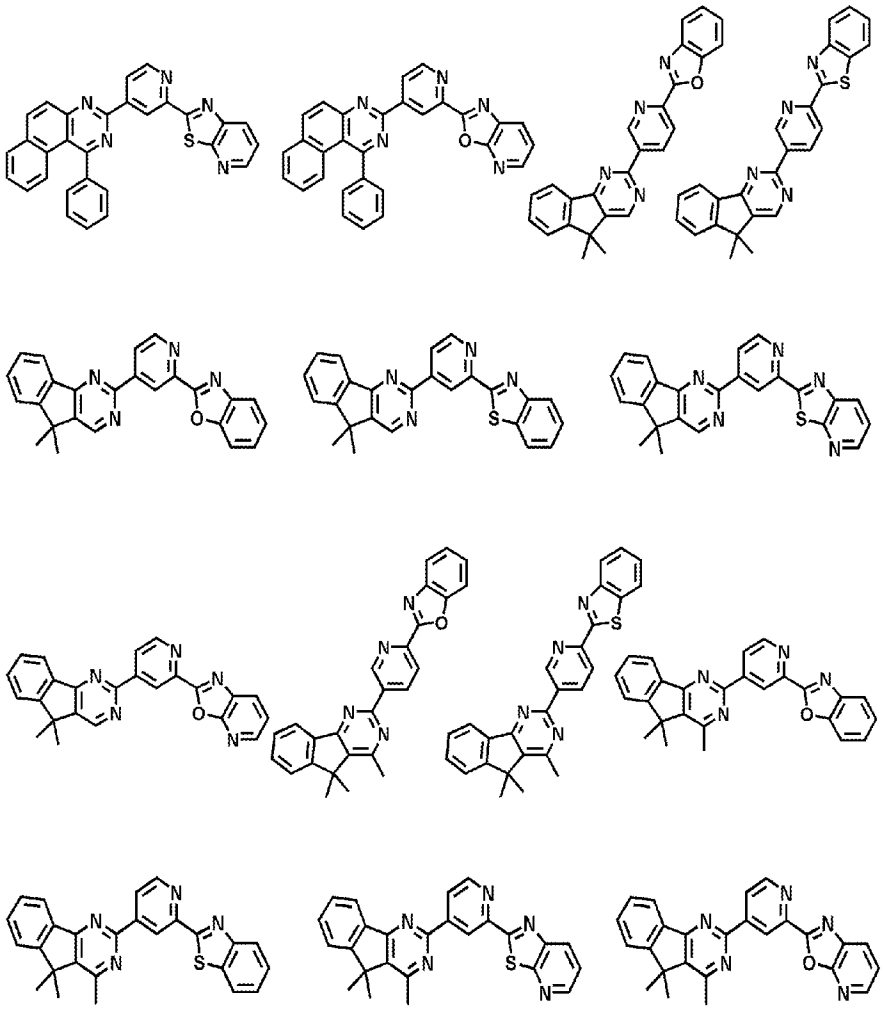


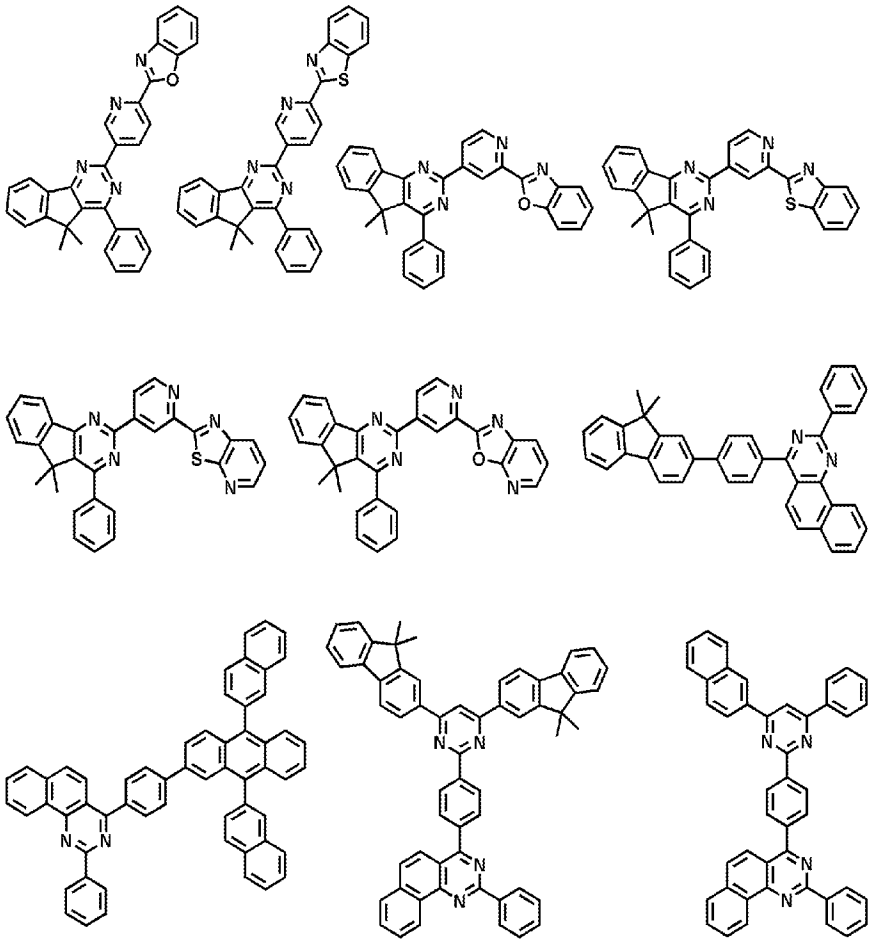


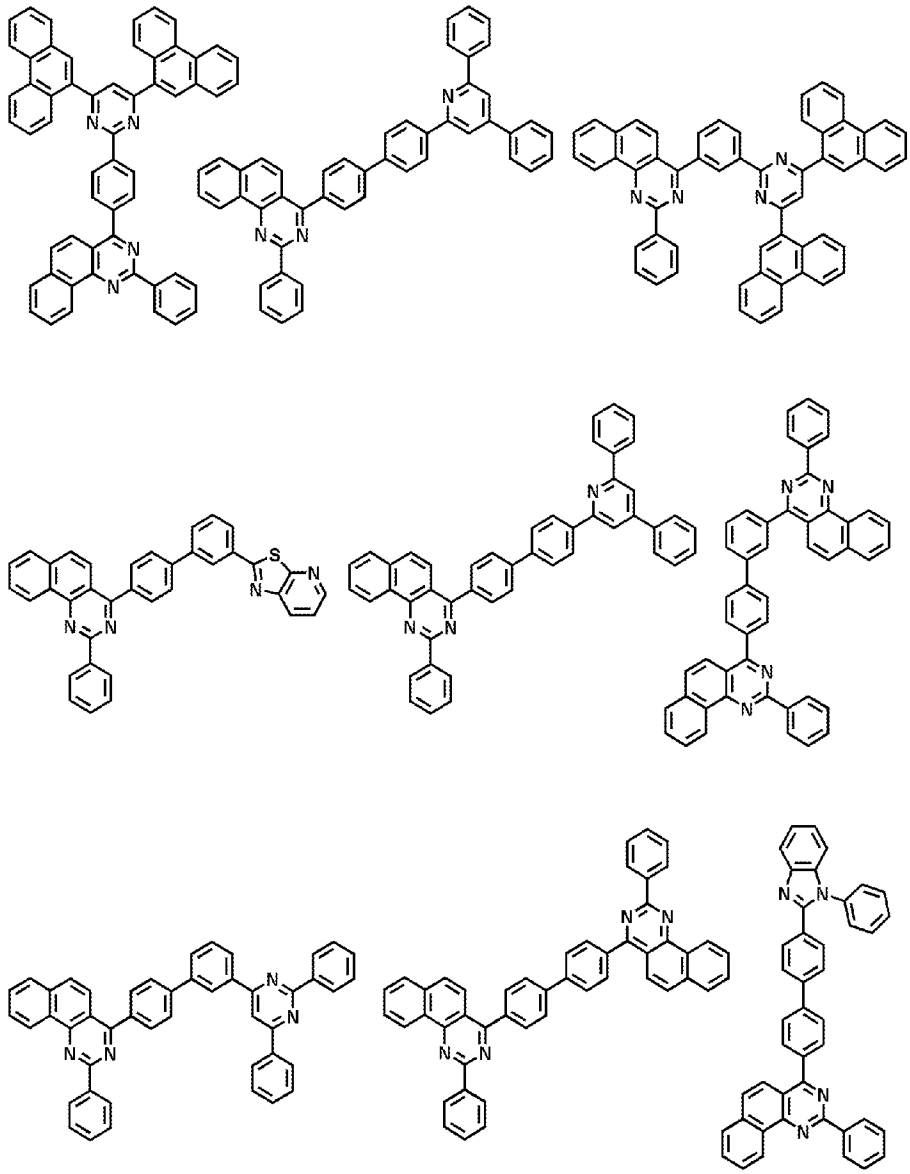


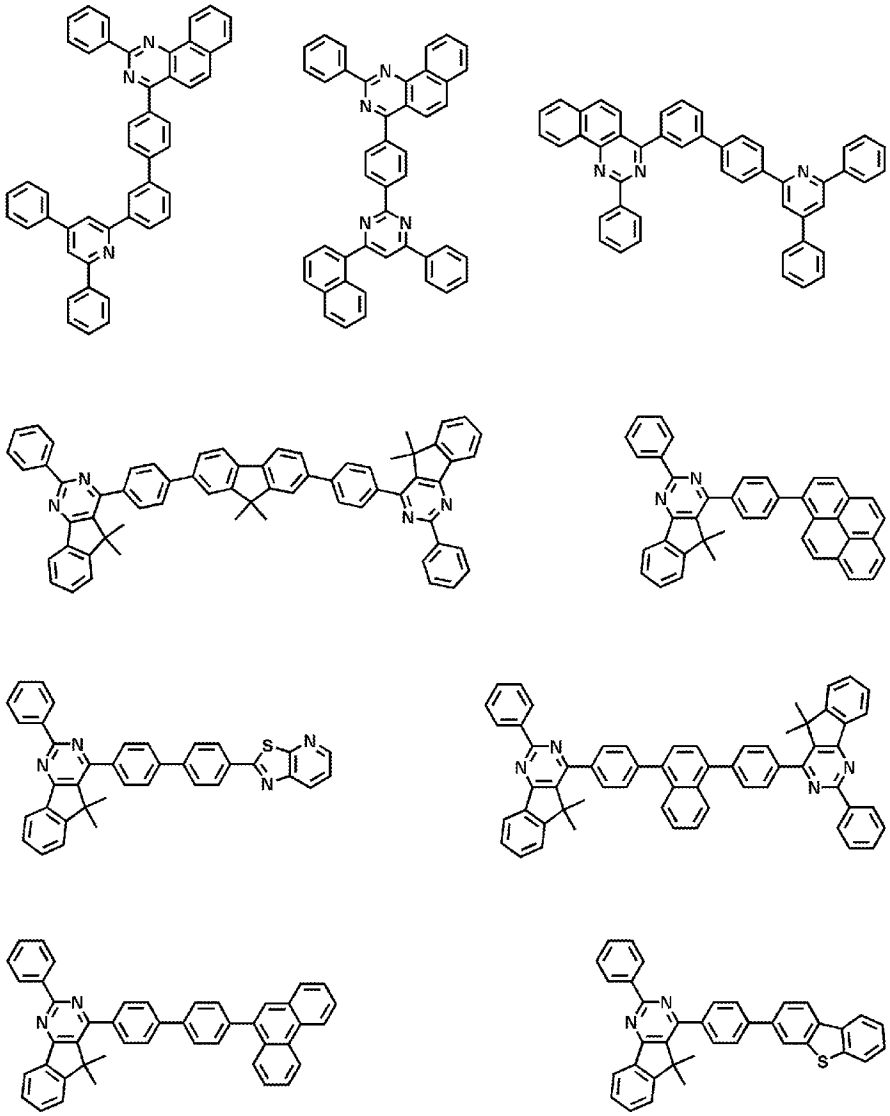


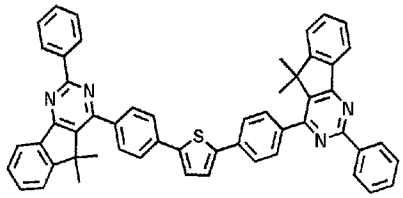


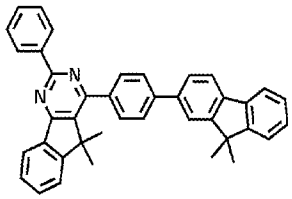


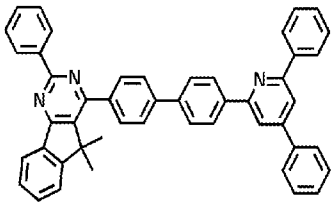


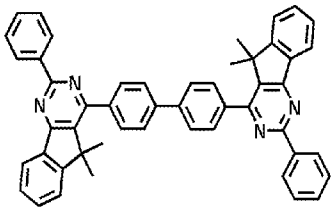


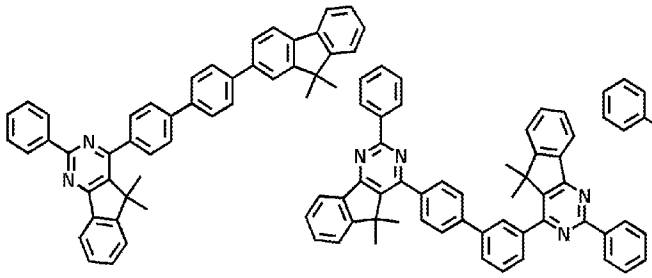


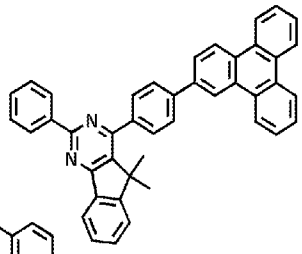


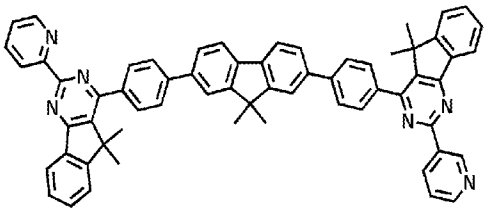


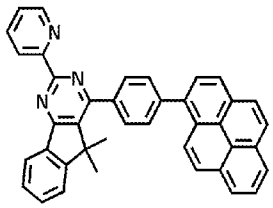


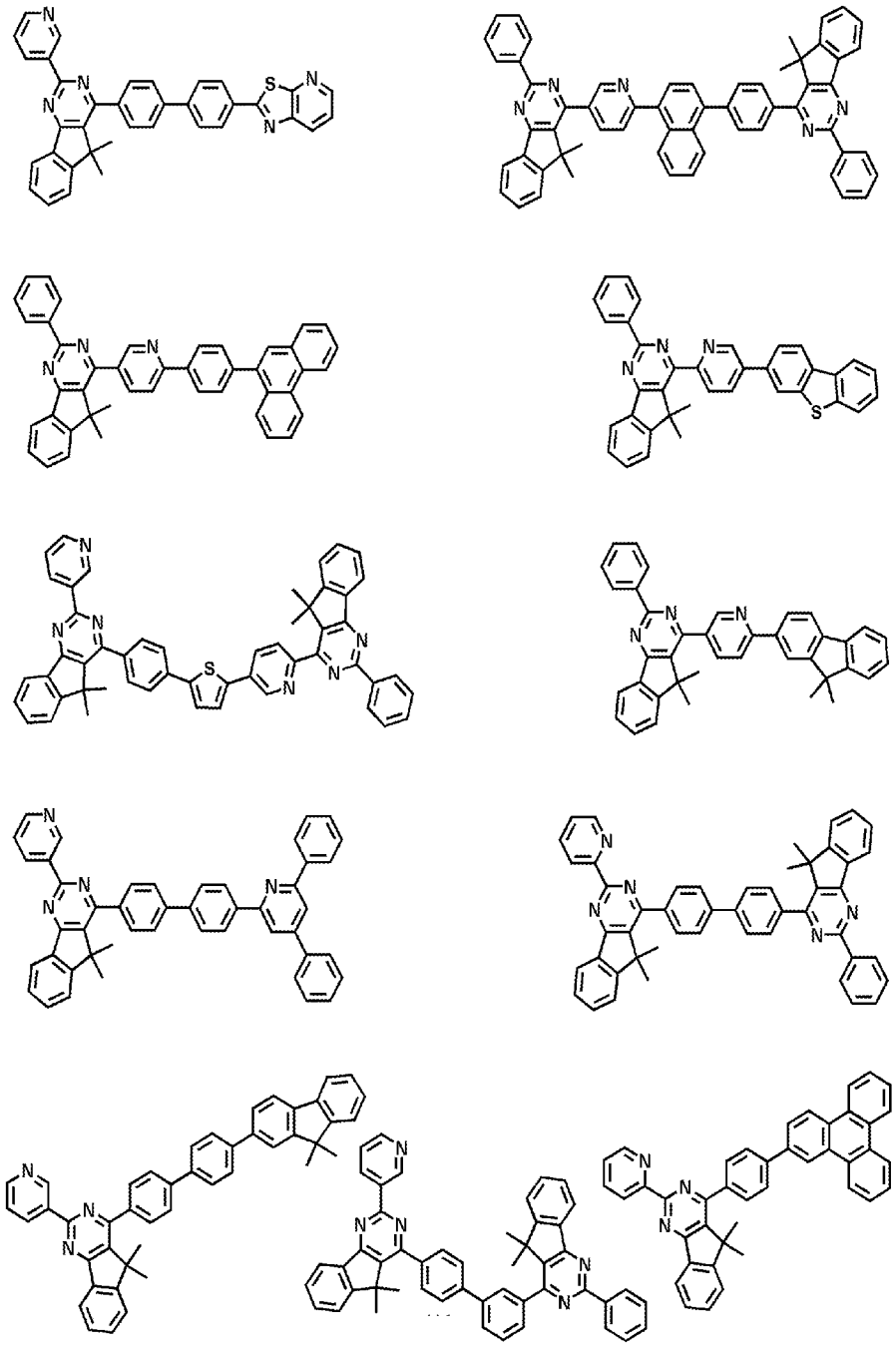








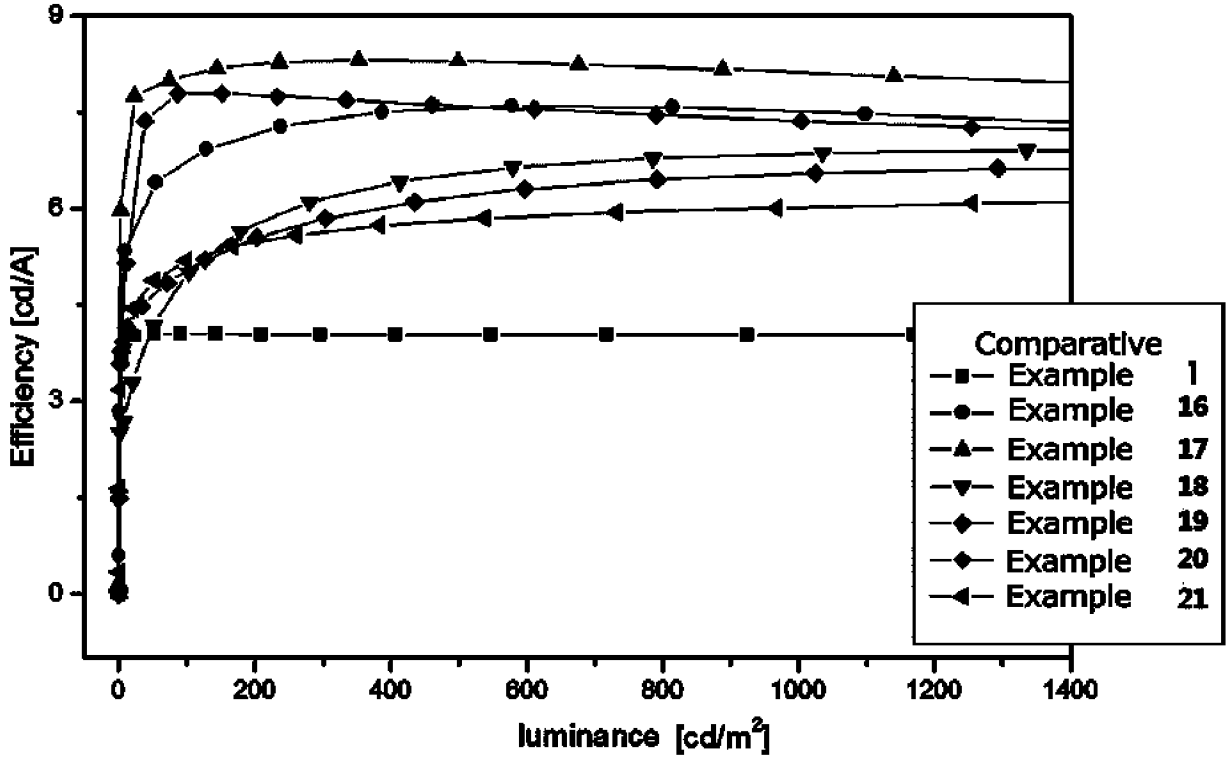




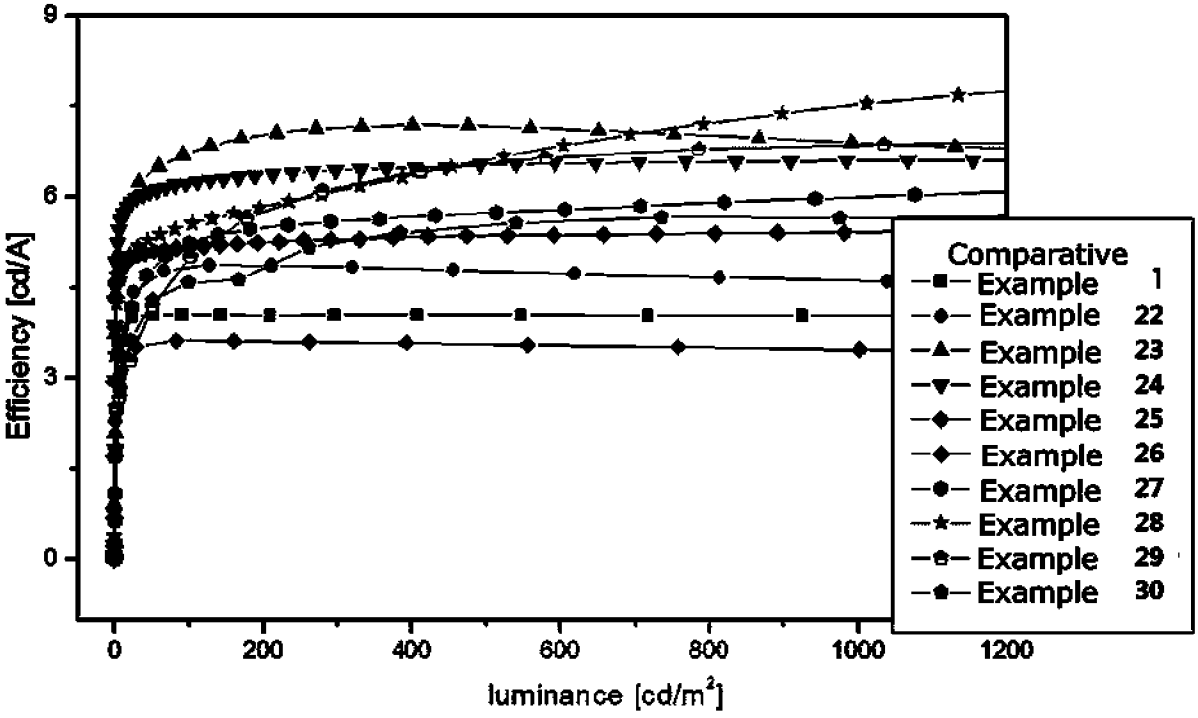
[Claim 6] An organic electroluminescent device containing the electron transport material of any one of claims 1 to 5.

[Claim 7] The organic electroluminescent device of claim 6, wherein it includes: a first electrode; a second electrode; and at least one organic layer interposed between the first and second electrodes, the organic layer including an electron transport layer in which the electron transport material is contained.

[Fig. 1]


Efficiency [cd/A]
luminance [cd/m²]
0
3
6
9
0
200
400
600
800
1000
1200
1400
Comparative Example 1
Example 16
Example 17
Example 18
Example 19
Example 20
Example 21

[Fig. 2]


Efficiency [cd/A]
luminance [cd/m²]
0
3
6
9
0
200
400
600
800
1000
1200
Comparative Example 1
Example 22
Example 23
Example 24
Example 25
Example 26
Example 27
Example 28
Example 29
Example 30

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2013/001371

A. CLASSIFICATION OF SUBJECT MATTER

C07D 239/74(2006.01)i, C07D 417/10(2006.01)i, C09K 11/06(2006.01)i, H01L 51/50(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C07D 239/74; H01L 51/00; C07D 239/26; C07D 403/04; C07D 401/04; C07D 401/10

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models
Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS(KIPO internal) & Keywords: "benzo[h]quinazoline", "benzo[f]quinazoline", "5,6-dihydrobenzo[h]quinazoline", "5,6-dihydrobenzo[f]quinazoline", "5H-indeno[1,2-d]pyrimidine", "9H-indeno[2,1-d]pyrimidine", "electroluminescent device"

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	MAMAEV, V. P. et al. Pyrimidines III. Dehydrogenation of 4-phenylbenzo [h]quinazoline derivatives. Khimiya Geterotsiklicheskikh Soedinenii. 1965, Vol. 1, No. 4, pp. 608-615 See page 410, compound VI.	1-5
X	ROBEV, S. K. New sysnthesis of quinazoline and benzoquinazoline derivatives. Tetrahedron Letters. 1983, Vol. 24, No. 40, pp. 4351-4354 See page 4352, compounds IVa-IVc, VIa-VIc.	1-5
X	ROBEV, S. K. 2,4-Disubstituted benzo/h/Quinazolines from N-(1-naphthyl)-amidines. Doklady Bolgarskoi Akademii Nauk. 1983, Vol. 36, No. 12, pp. 1551-1553 See compounds IIa-IIc, Va-Ve.	1-5
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☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

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

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
14 May 2013 (14.05.2013)	**15 May 2013 (15.05.2013)**
Name and mailing address of the ISA/KR Korean Intellectual Property Office 189 Cheongsa-ro, Seo-gu, Daejeon Metropolitan City, 302-701, Republic of Korea Facsimile No. 82-42-472-7140	Authorized officer KIM, Eun Young Telephone No. -

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2013/001371

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2004-039786 A1 (CIBA SPECIALTY CHEMICALS HOLDING INC. et al.) 13 May 2004 See abstract; example 15; claims 1,2,9	1-7
X	LEONETTI, F. et al. Design, Snthesis, and 3D QSAR of Novel Potent and Selective Aromatase Inhibitors. Journal of Medicinal Chemistry. 2004, Vol. 47, pp. 6792-6803 See scheme 1, compounds 4 and 6.	1-4
X	WO 93-14080 A1 (E.I. DU PONT DE NEMOURS AND COMPANY et al.) 22 July 1993 See index table A, compounds 3-7, 12, 14-19.	1-4
X	CH 599569 A5 (HOECHST AG) 31 May 1978 See table 1, 4a and 4b; column 7, formula 4.	1
X	MIYASHI, T. et al. The Intermolecular Nitrene-Type 1,1-Cycloaddition Reaction of Allyl-Substituted Diazomethanes. Journal of American Chemical Society. 1986, Vol. 108, pp. 1617-1632 See scheme IV, compounds 16 and 17.	1
X	CHAO, B. et al. Copper(I)-Mediated Cascade Reactions: An Efficient Approach to the Synthesis of Functionalized Benzofuro[3,2-d]pyrimidines. Organic Letters. 26 April 2012 (published on web), Vol. 12, No. 9, pp. 2398-2401 See table 1, compound 10a; table 2, compounds 10b, 10h-10j and 10l.	1,2
X	CHETONI, F. et al. Synthesis of Novel 1-Aryl[1]benzoxepino[5,4-c]pyrazole and [1]benzoxepino[5,4-d]pyrimidine Derivatives. Journal of Heterocyclic Chemistry. 1993, Vol. 30, pp. 1653-1658 See scheme 1, table 7, compounds 27a, 28a, 31b, 32b, 35c and 36c.	1,2
X	ROBBA, M. et al. Synthese de benzo (1) thieno [2,3-d] pyrimidines et de benzo (1) thieno [3,2-d] pyrimidines. Tetrahedron Letters. 1972, Vol. 13, No. 44, pp. 4549-4551 See page 4550, compound 6.	1,2
X	KUMAR, A. S. et al. Synthesis of pyrido[2,3-b]indoles and pyrimidoindoles via Pd-catalyzed amidation and cyclization. Organic & Biomolecular Chemistry. 25 April 2012 (published on web), Vol. 10, No. 26, pp. 5084-5093 See table 4, compounds 3a-3g, 3i-3m and 3o-3t.	1,2
PX	KR 10-2012-0117693 A (SFC CO., LTD) 24 October 2012 See abstract; claims 1, 3-6; compounds of formula 505, 510, 512, 517-522, 525, 527-529, 532-539, 548-550, 559, 560, 563, 564, 570, 573, 579, 581, 584, 585, 597-599, 609, 610, 615.	1-7

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR2013/001371

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KR 10-2012-0117693 A	24.10.2012	None	