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(12) **United States Patent**
Suzuki et al.(10) **Patent No.:** **US 6,994,922 B2**
(45) **Date of Patent:** **Feb. 7, 2006**(54) **ORGANIC LUMINESCENCE DEVICE WITH
A FUSED POLYNUCLEAR COMPOUND**(75) Inventors: **Koichi Suzuki**, Kanagawa (JP);
Akihiro Senoo, Kanagawa (JP);
Hiroshi Tanabe, Kanagawa (JP)(73) Assignee: **Canon Kabushiki Kaisha**, Tokyo (JP)(*) Notice: Subject to any disclaimer, the term of this
patent is extended or adjusted under 35
U.S.C. 154(b) by 16 days.(21) Appl. No.: **10/940,734**(22) Filed: **Sep. 15, 2004**(65) **Prior Publication Data**

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Related U.S. Application Data(62) Division of application No. 10/077,800, filed on Feb.
20, 2002, now Pat. No. 6,830,829.(30) **Foreign Application Priority Data**

Feb. 22, 2001 (JP) 2001-046225

Feb. 14, 2002 (JP) 2002-036804

(51) **Int. Cl.**
H05B 33/14 (2006.01)(52) **U.S. Cl.** **428/690**; 428/917; 313/504;
313/506(58) **Field of Classification Search** 428/690,
428/917; 313/504, 506; 525/301.16
See application file for complete search history.(56) **References Cited****U.S. PATENT DOCUMENTS**

4,539,507 A	9/1985	VanSlyke et al.	313/504
4,720,432 A	1/1988	VanSlyke et al.	428/457
4,885,211 A	12/1989	Tang et al.	428/457
5,077,142 A	12/1991	Sakon et al.	428/690
5,130,603 A	7/1992	Tokailin et al.	313/504
5,151,629 A	9/1992	VanSlyke	313/504
5,227,252 A	7/1993	Murayama et al.	428/690
5,247,190 A	9/1993	Friend et al.	257/40
5,317,169 A	5/1994	Nakano et al.	257/40
5,382,477 A	1/1995	Saito et al.	428/690
5,409,783 A	4/1995	Tang et al.	428/690
5,514,878 A	5/1996	Holmes et al.	257/40
5,536,949 A	7/1996	Hosokawa et al.	257/40
5,635,308 A	6/1997	Inoue et al.	428/690
5,645,948 A	7/1997	Shi et al.	428/690
5,672,678 A	9/1997	Holmes et al.	528/373
5,726,457 A	3/1998	Nakano et al.	257/40
5,766,779 A	6/1998	Shi et al.	428/690
6,093,864 A	7/2000	Tokailin et al.	585/25
6,203,933 B1	3/2001	Nakaya et al.	428/690
6,596,415 B2	7/2003	Shi et al.	428/690
6,635,364 B1	10/2003	Igarashi	428/690
6,830,829 B2 *	12/2004	Suzuki et al.	428/690

FOREIGN PATENT DOCUMENTS

JP 2-247278 10/1990

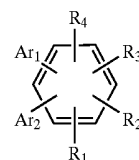
JP	3-255190	11/1991
JP	4-68076	3/1992
JP	4-145192	5/1992
JP	5-32966	2/1993
JP	5-202356	8/1993
JP	5-247460	9/1993
JP	6-228552	8/1994
JP	6-240244	8/1994
JP	7-109454	4/1995
JP	8-311442	11/1996
JP	9-202878	8/1997
JP	9-227576	9/1997
JP	9-241629	9/1997
JP	2000-26334	1/2000
JP	2000-268964	9/2000
JP	2001-192651	7/2001

OTHER PUBLICATIONSNorio Miyaura et al., "Palladium-Catalyzed Cross-Coupling
Reactions of Organoboron Compounds," 95(7) *Chem. Rev.*
2457-2483 (1995).Takakazu Yamamoto et al., "A Novel Type of Polycondensa-
tion Utilizing Transition Metal-Catalyzed C-C Coupling—I.
Preparation of Thermostable Polyphenylene Type
Polymers," 51(7) *Bull. Chem. Soc. Jpn.* 2091-2097 (1978).

(Continued)

Primary Examiner—Marie Yamnitzky(74) *Attorney, Agent, or Firm*—Fitzpatrick, Cella, Harper &
Scinto(57) **ABSTRACT**

An organic luminescence device having an anode and a cathode and at least one organic luminescence function layer disposed between the anode and the cathode. At least one organic luminescence function layer described above includes a layer of a fused polynuclear compound represented by the following formula (I):



(I)

wherein R₁, R₂, R₃ and R₄ independently denote hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or cyano group; and Ar₁ and Ar₂ independently denote a substituted or unsubstituted fused polynuclear aromatic group or a substituted or unsubstituted fused polynuclear heterocyclic group.

OTHER PUBLICATIONS

F.J. Weigert, "HZSM-5-Catalyzed Isomerization of Alkylanilines," 52(15) *J. Org. Chem.* 3296-3298 (1987).

C.W. Tang et al., "Organic Luminescent Diodes," 51(12) *Appl. Phys. Lett.* 913-915 (Sep. 1987).

J.H. Burroughes et al., "Light-Emitting Diodes Based on Conjugated Polymers," 347 *Nature* 539-541 (Oct. 1990).

Yasuhiko Shirota et al., "Development of Hole-Blocking Amorphous Molecular Materials and Their Application in Organic Light-Emitting Diodes," in *Organic Light-Emitting Materials, Proceedings of SPIE*, vol. 4464, pp. 203-210 (2002).

* cited by examiner

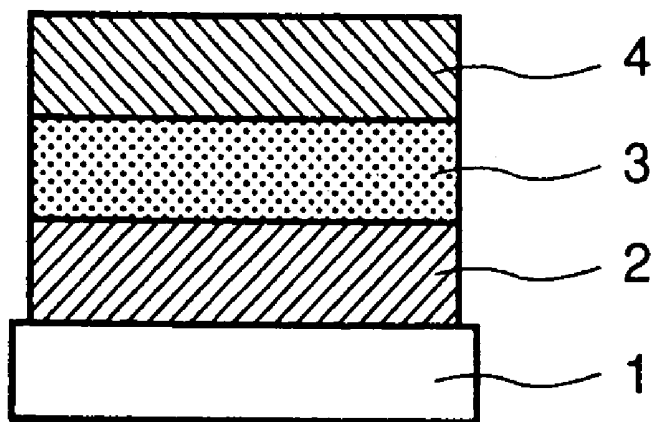


FIG. 1

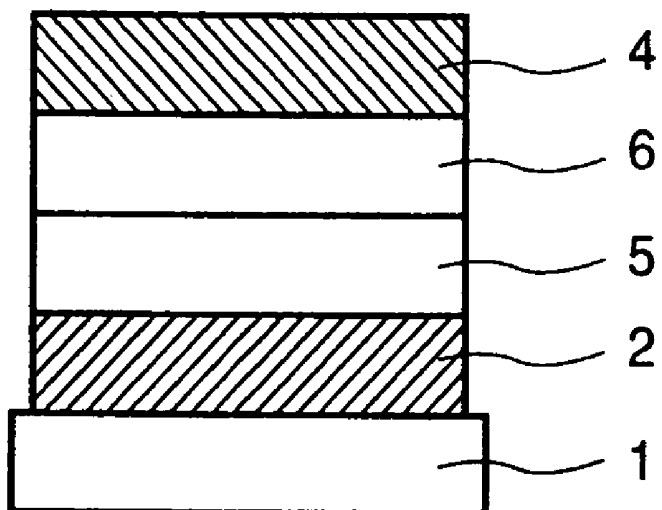


FIG. 2

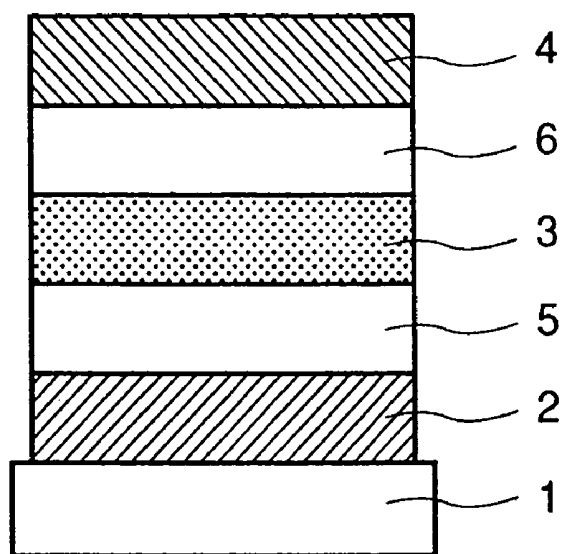


FIG. 3

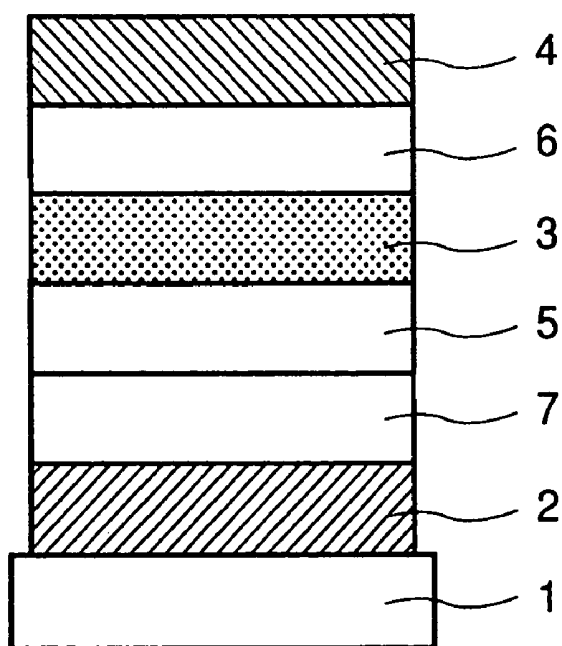


FIG. 4

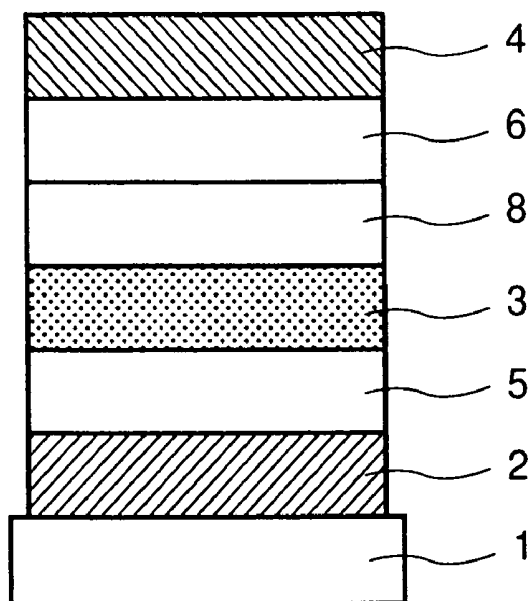


FIG. 5

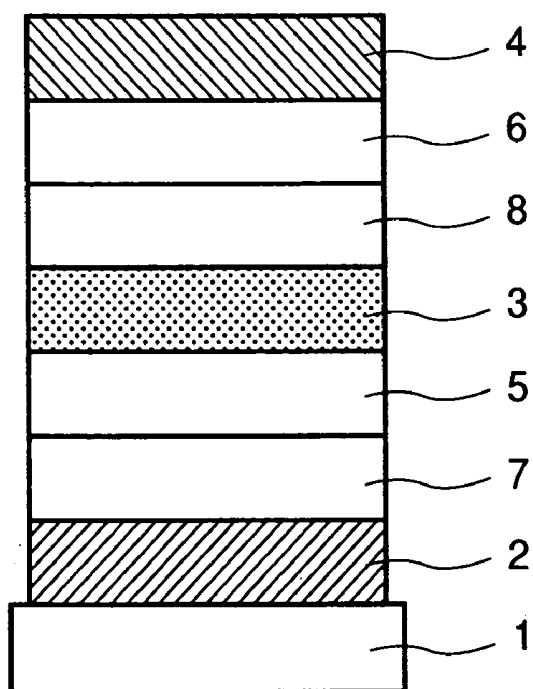


FIG. 6

ORGANIC LUMINESCENCE DEVICE WITH A FUSED POLYNUCLEAR COMPOUND

This application is a division of application Ser. No. 10/077,800, filed Feb. 20, 2002, now U.S. Pat. No. 6,830,829, which is incorporated herein by reference.

FIELD OF THE INVENTION AND RELATED ART

The present invention relates to a specific fused polynuclear (or polycyclic) compound and an organic luminescence device using the fused polynuclear compound. An organic luminescence device is a device such that a thin film containing a fluorescent organic compound is sandwiched between an anode and a cathode, and holes and electrons are injected from the anode and the cathode, respectively, into the fluorescent organic compound layer to generate excitons, which emit light at the time of being returned to a ground state.

More specifically, according to Kodak's study (Appl. Phys. Lett., 51,913 (1987)), when a voltage of ca. 10 volts is applied to a function separation-type organic luminescence device including two layers of an aluminum quinolinol complex (as electron transport and luminescence material) and a triphenylamine derivative (as hole transport material) disposed between an anode of ITO (indium tin oxide) and a cathode of a magnesium-silver alloy, an emission luminance of ca. 1000 cd/m² is obtained. Related patents thereof are, e.g., U.S. Pat. Nos. 4,539,507; 4,720,432; and 4,885,211.

Further, it is impossible to effect luminescence ranging from ultraviolet region to infrared region by appropriately changing the species of a fluorescent organic compound used. In recent years, studies on various fluorescent organic compounds have been made extensively as, e.g., described in U.S. Pat. Nos. 5,151,629 5,409,783; and 5,382,477; and Japanese Laid-Open Patent Application (JP-A) Nos. 2-247278 (corr. to U.S. Pat. Nos. 5,130,603 and 6,093,864); 3-255190 (corr. to U.S. Pat. No. 5,227,252); 5-202356; 9-202878; and 9-227576.

In addition to the above-mentioned organic luminescence devices using low-molecular weight materials, an organic luminescence device using a conjugated system polymer has been reported by a research group of Kenbridge University ("Nature", 347,539 (1990)). According to this report, by forming a single layer of PPV (polyphenylenevinylene) by means of wet coating, luminescence from the layer has been confirmed.

Related patents as to organic luminescence devices using conjugated system polymers may include: U.S. Pat. Nos. 5,247,190; 5,514,878; and 5,672,678; JP-A 4-145192 (corr. to U.S. Pat. Nos. 5,317,169 and 5,726,457) and JP-A 5-247460.

As described above, recent progress of the organic luminescence devices is noticeable. More specifically, it is possible to realize a thin and light-weight luminescence device allowing high luminance at low applied voltage, variety of emission wavelength and high-speed responsiveness, thus suggesting possibilities of application to various uses.

However, the organic luminescence devices are required to exhibit further improved performances such as light output at high luminance and high conversion efficiency. Further, the organic luminescence devices have been accompanied with problems in terms of durability such as a change in luminance with time due to continuous use for a long time, and a deterioration (e.g., an occurrence of dark spots

due to leakage of current) by ambient gas containing oxygen or by humidity. Moreover, in the case where the organic luminescence devices are applied to full-color displays, it is necessary to realize luminance of red (R), green (G) and blue (B) with good color purities but resultant color purities have been insufficient as yet.

As fluorescent organic compounds for use in an electron transport layer and/or a luminescence layer, a large number of aromatic compounds or fused polynuclear aromatic compounds have been proposed as described in, e.g., JP-A 4-68076, 5-32966, 6-228552, 6-240244, 7-109454, 8-311442 (corr. to U.S. Pat. No. 6,203,933), 9-241629, 2000-26334, and 2000-268964. However, resultant emission luminances and durabilities have been still insufficient.

SUMMARY OF THE INVENTION

A principal object of the present invention is to provide an organic luminescence device having solved the above-mentioned problems.

A specific object of the present invention is to provide an organic luminescence device capable of effecting output of light with very high efficiency and luminance by using a specific fused polynuclear aromatic compound.

Another object of the present invention is to provide an organic luminescence device having a high durability.

A further object of the present invention is to provide an organic luminescence device which can be prepared readily and relatively inexpensively.

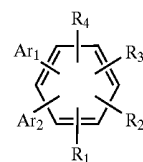
According to the present invention, there is provided an organic luminescence device, comprising:

a pair of an anode and a cathode, and

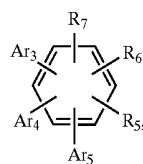
at least one organic luminescence function layer disposed between the anode and the cathode, wherein

at least one organic luminescence function layer described above comprises a layer comprising at least one species of a fused polynuclear compound characterized by a substituted benzene structure having at least two fused polynuclear aromatic or heterocyclic groups as substituents.

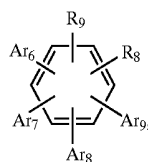
The fused polynuclear compound used in the present invention is represented by any one of the following formulas (I) to (VII):



(I)



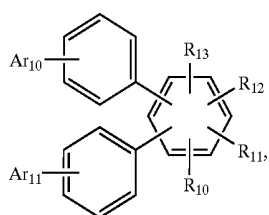
(II)



(III)

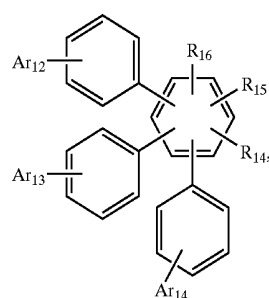
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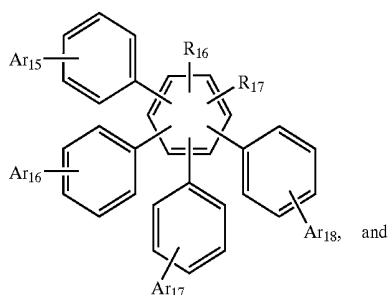
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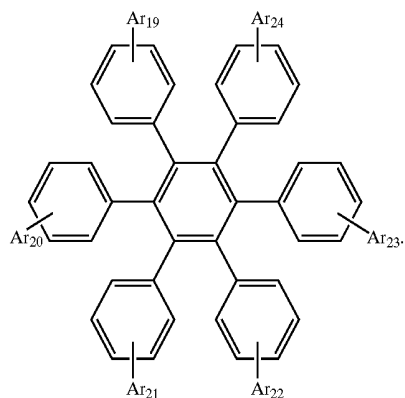
(V)

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(VI)

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(VII)

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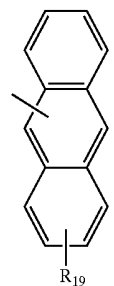
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In the above formulas (I) to (VII), R₁ to R₁₈ independently denote hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or cyano group; and Ar₁ to Ar₂₄ independently denote a substituted or unsubstituted fused polynuclear aromatic group or a substituted or unsubstituted fused polynuclear heterocyclic group.

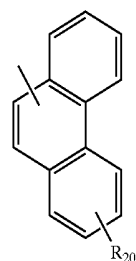
In the present invention, Ar₁ to Ar₂₄ in the above-mentioned formulas (I) to (VII) may preferably denote a substituted or unsubstituted fused polynuclear group comprising at least three fused benzene rings or at least four fused benzene rings, more preferably be a fused polynuclear group represented by any one of the following formulas (VIII) to (XIII).

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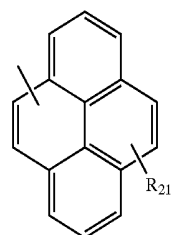
(VIII)



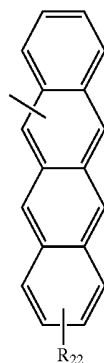
(IX)



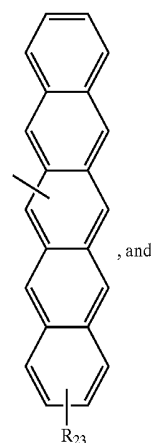
(X)



(XI)

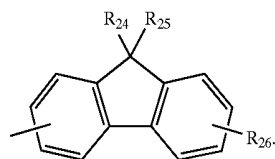


(XII)



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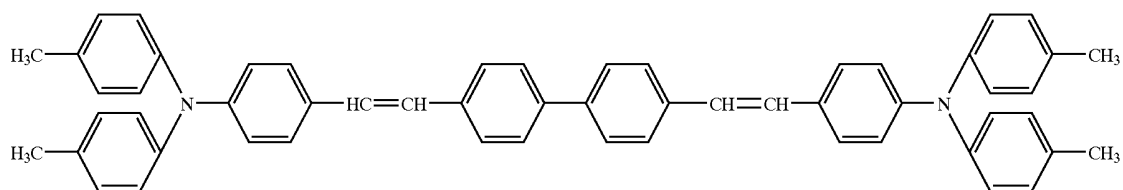
In the above formulas (VIII) to (XIII), R₁ to R₂₆ independently denote hydrogen atom, an aralkyl group, a substituted or unsubstituted aralkyl group, a substituted or

(XIII)

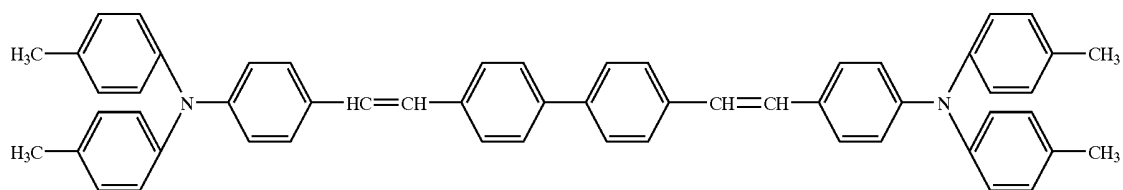
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unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or cyano group.

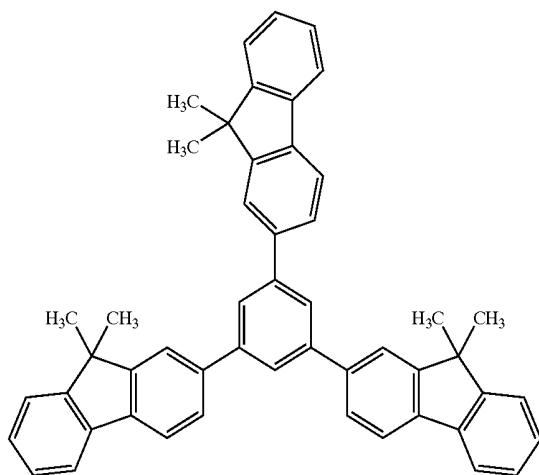
In the present invention, when the above-mentioned at least one organic luminescence function layer comprises an electron transport layer and or a is luminescence layer, the electron transport layer or luminescence layer may preferably comprise a fused polynuclear compound represented by any one of the formulas (I) to (VII). In this case, in a more preferred embodiment, the luminescence layer comprises a fused polynuclear compound represented by any one of the formulas (I) to (VII) and a compound represented by the following formula:



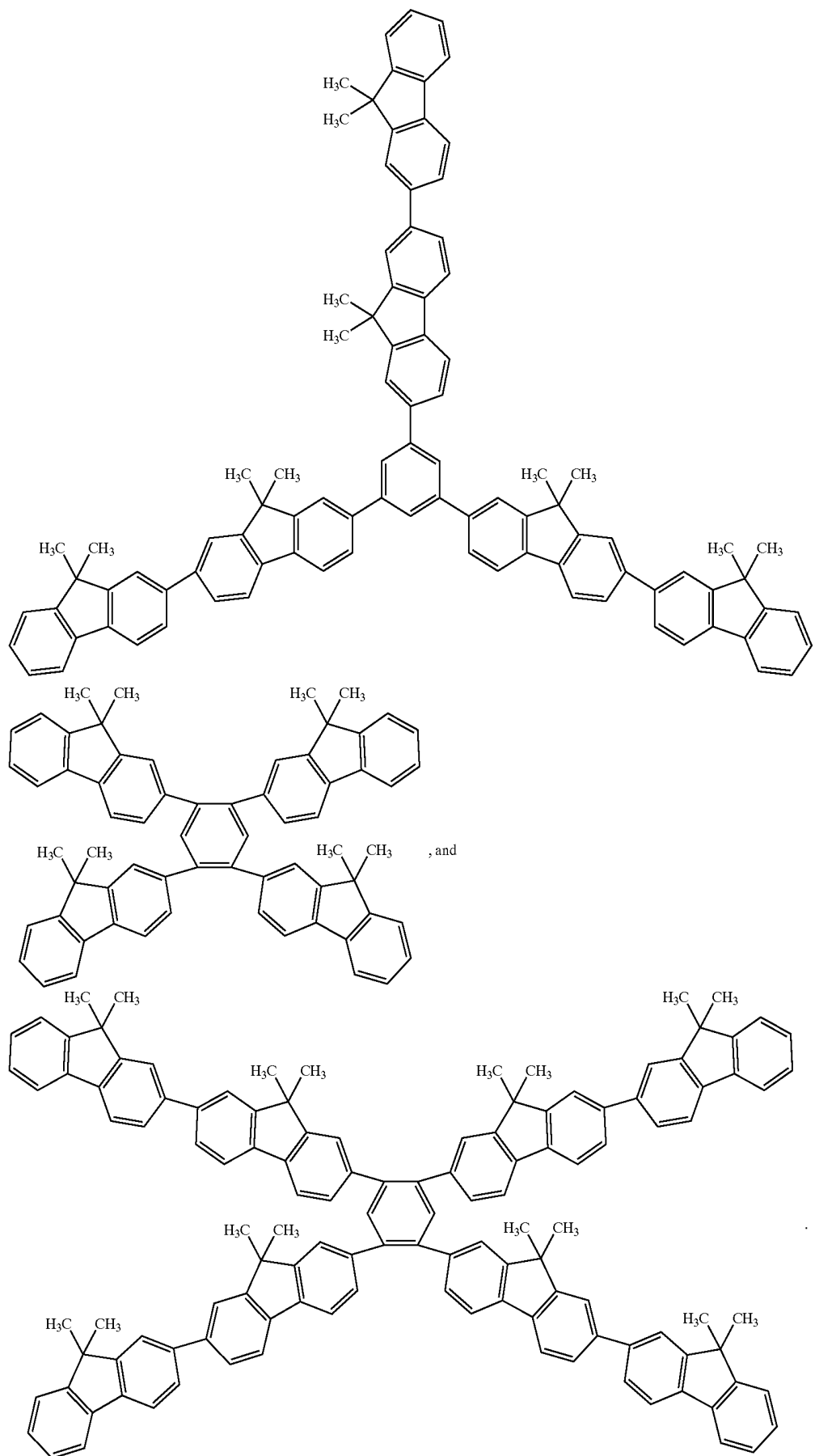
Further, when the above-mentioned at least one organic luminescence function layer comprises an electron transport layer and or a luminescence layer, the electron transport layer may preferably comprise a hole transporting material and a yellow luminescent material, and said luminescence later may preferably comprise a fused polynuclear compound represented by any one of the formulas (I) to (VII) and a compound represented by the following formula:



The fused polynuclear compound used in the present invention may preferably be represented by any one of the following formulas:



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These and other objects, features and advantages of the present invention will become more apparent upon a consideration of the following description of the preferred embodiments of the present invention taken in conjunction with the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

FIGS. 1 to 6 are respectively a schematic sectional view showing an embodiment of the organic luminescence device according to the present invention.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Hereinbelow, the present invention will be described more specifically.

The organic luminescence device according to the present invention comprises a pair of electrodes composed of an anode and a cathode and at least one organic luminescence function layer disposed between the pair of electrodes. Examples of such an organic luminescence function layer may include a luminescence layer, a hole transport layer, an electron transport layer, a hole injection layer and a hole/exciton blocking layer.

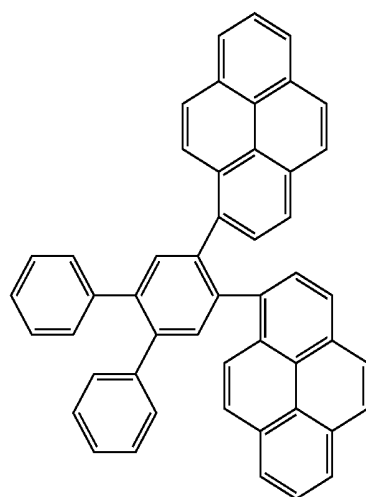
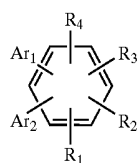
In the present invention, the above-mentioned at least one organic luminescence function layer may preferably have one to five layer structures specifically described hereinafter with reference to FIGS. 1-6.

At least one organic luminescence function layer described above includes at least one layer comprising a fused polynuclear compound represented by any one of the above-mentioned formulas (I) to (VII).

Specific examples of R_1 to R_{26} and Ar_1 to Ar_{24} in the formulas (I) to (VII) for the fused polynuclear compound used in the present invention will be shown below.

[For R_1 to R_{24}]

Examples of alkyl group may include methyl, ethyl, n-propyl, iso-propyl, n-butyl, tert-butyl and octyl.



Examples of aralkyl group may include benzyl and phenethyl.

Examples of aryl group may include phenyl, biphenyl and terphenyl.

5 Examples of heterocyclic group may include thienyl, pyrrolyl, pyridyl, quinolyl, carbazolyl, oxazolyl, oxadiazolyl, thiazolyl, thiadiazolyl, terthienyl and terpyrrolyl.

Examples of substituted amino group may include dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino and dianisolylamino.

10 R_1 to R_{26} may be hydrogen atom or cyano group as described above.

Herein, the terms "aryl group" and "heterocyclic group" for R_1 to R_{26} mean those free from a fused ring structure as in those for Ar_1 to Ar_{24} .

15 [For Ar_1 to Ar_{24}]

Examples of fused polynuclear aromatic group may include naphthyl, anthryl, phenanthryl, pyrenyl, tetracenyl, pentacenyl, fluorenyl, triphenylenyl and peryleryl, more preferably those represented by the above-mentioned formulas (VIII) to (XIII).

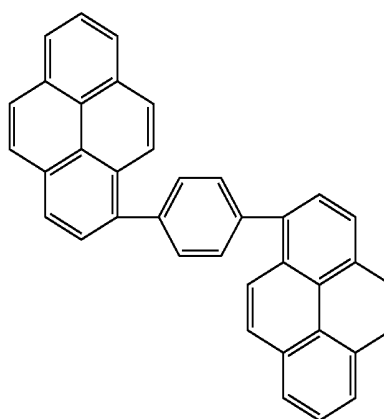
20 Examples of fused polynuclear heterocyclic group may include acridinyl and fluorenonyl.

Examples of substituents for the above-mentioned groups for R_1 to R_{26} and Ar_1 to Ar_{24} may include: alkyl groups such as methyl, ethyl and propyl; aralkyl groups such as benzyl and phenethyl; aryl groups such as phenyl, naphthyl, anthryl, phenanthryl, pyrenyl, tetracenyl, pentacenyl, and fluorenyl; heterocyclic groups such as thienyl, pyrrolyl and pyridyl; amino groups such as dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino and dianisolylamino; alkoxy groups such as methoxy, ethoxy, propoxy and phenoxy; cyano group; and nitro group.

35 Hereinafter, the fused polynuclear compounds of the formulas (I) to (VII) used in the present invention will be shown specifically by their structural formulas (Example Compound Nos. 1 to 66) for the respective formulas (I) to (VII) but are not restricted to the following specific compounds.

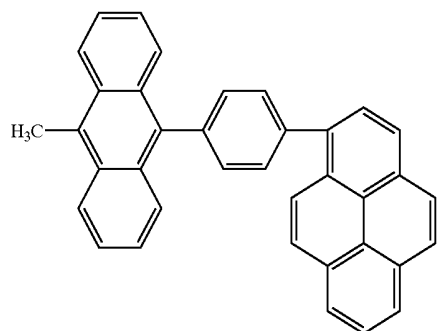
Formula [I]

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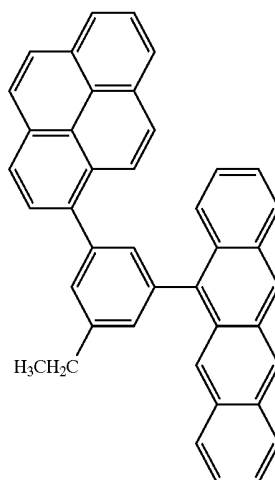


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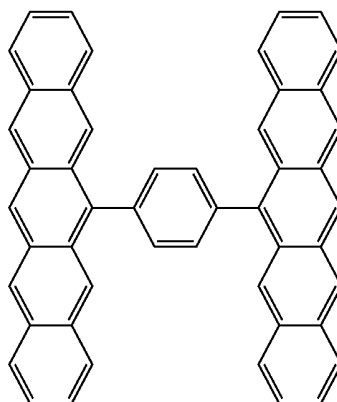
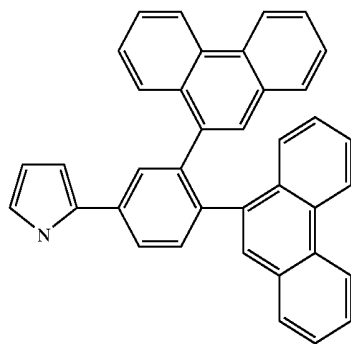
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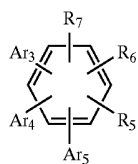
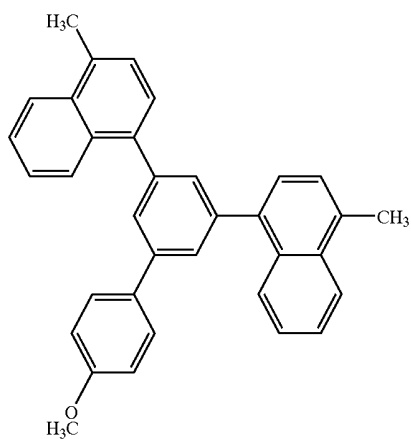
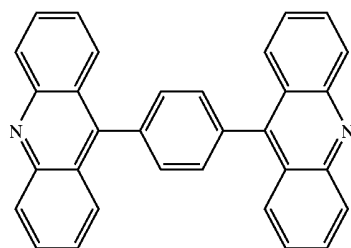
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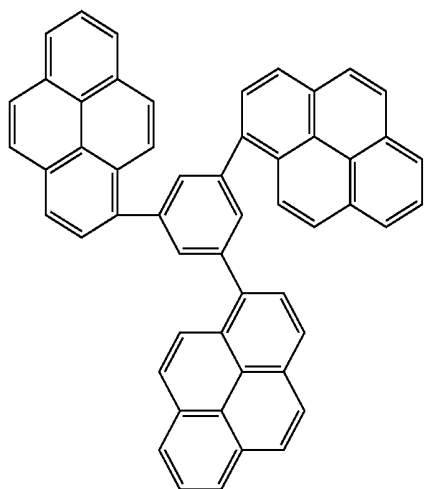
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Formula [II]

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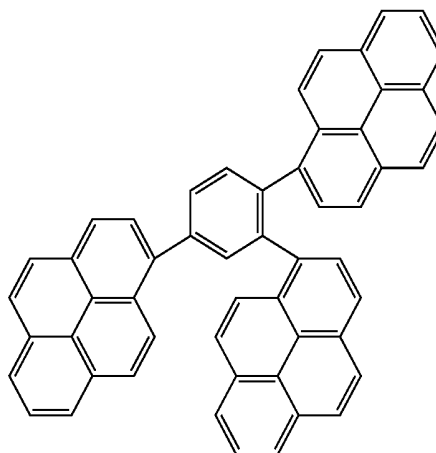


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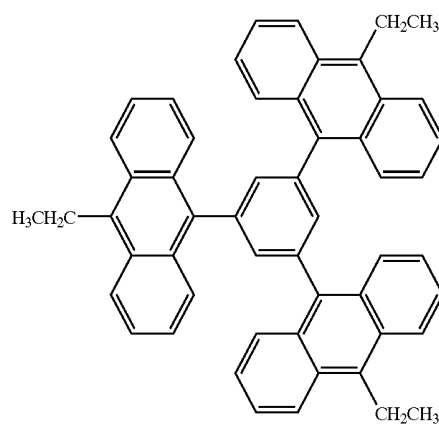
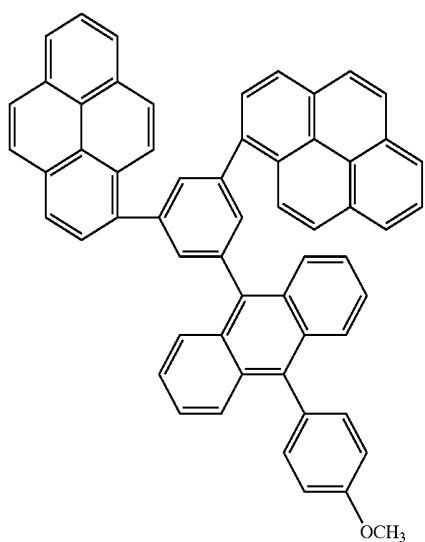
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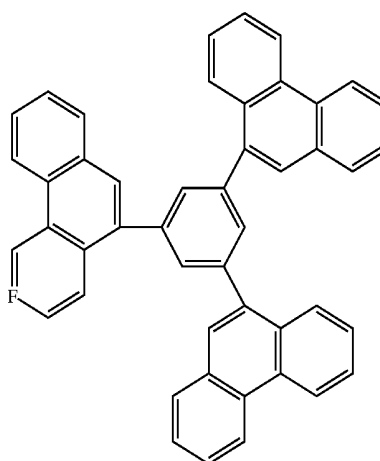
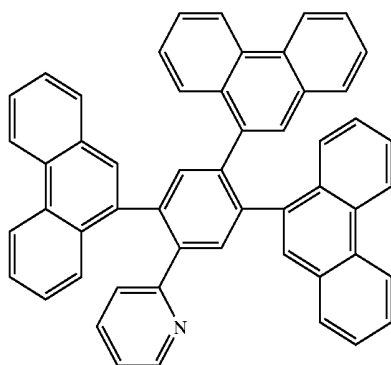
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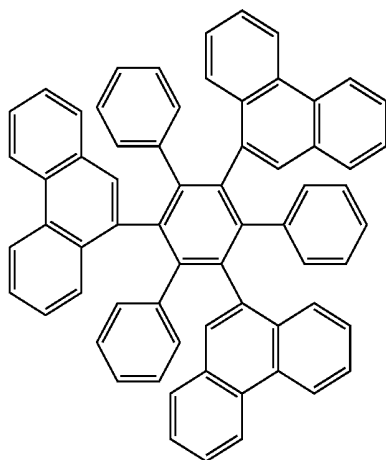


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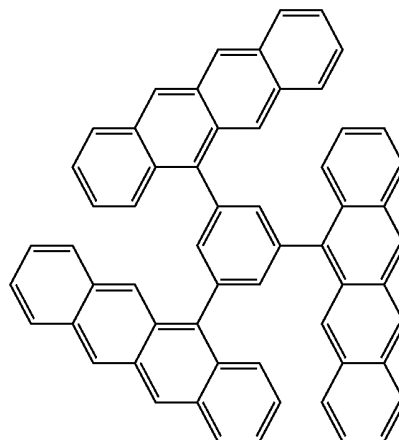
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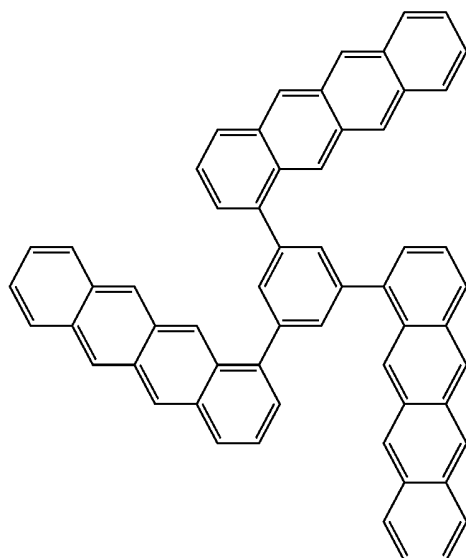


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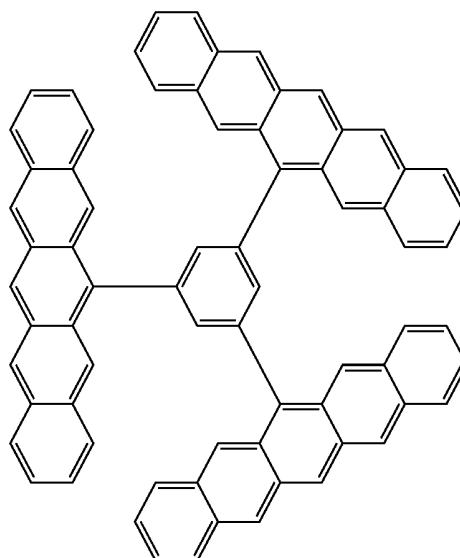
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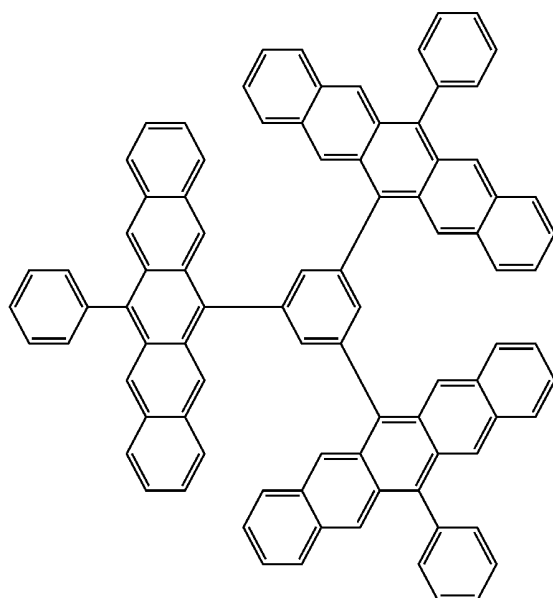
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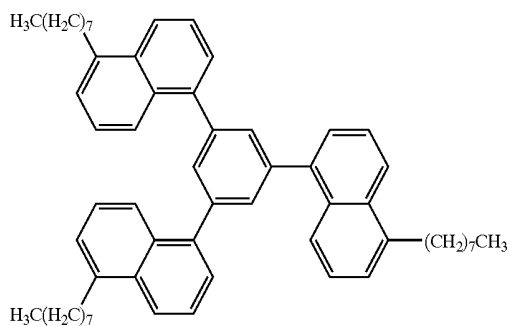
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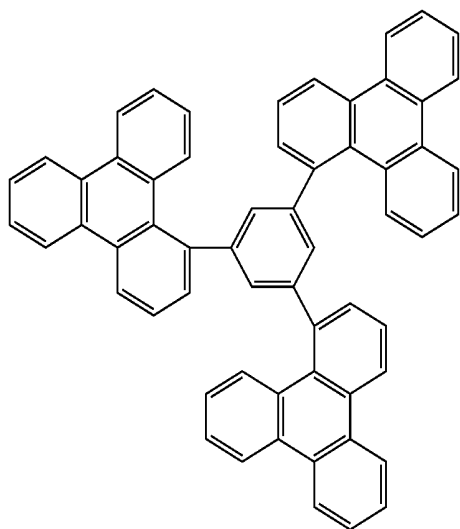
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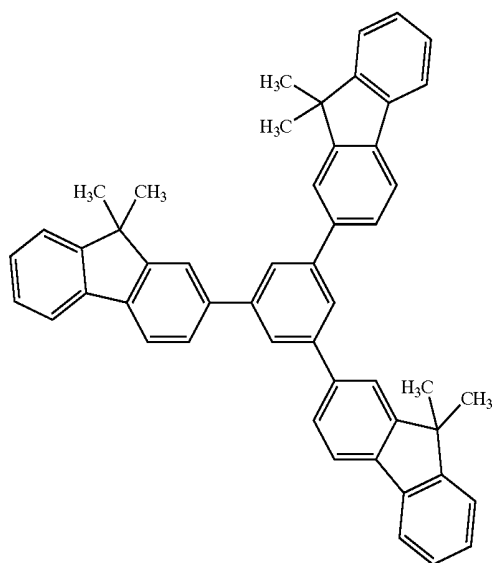


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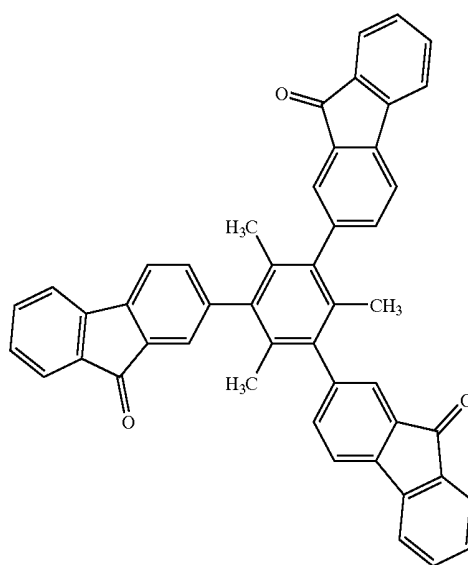
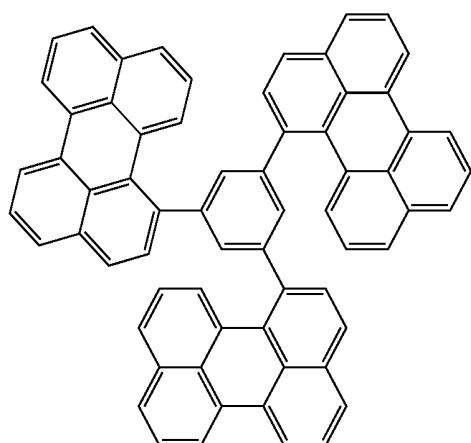
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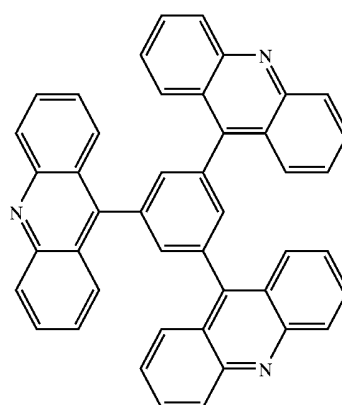
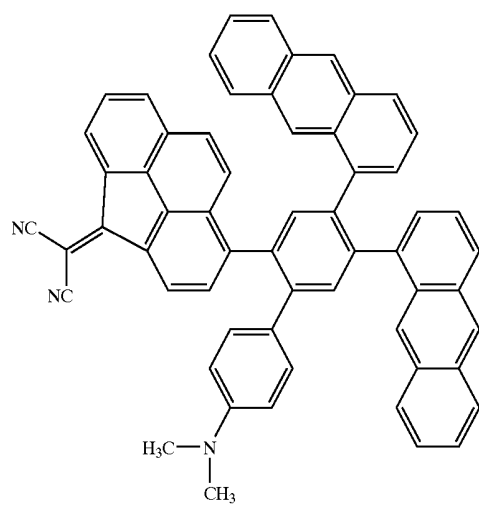
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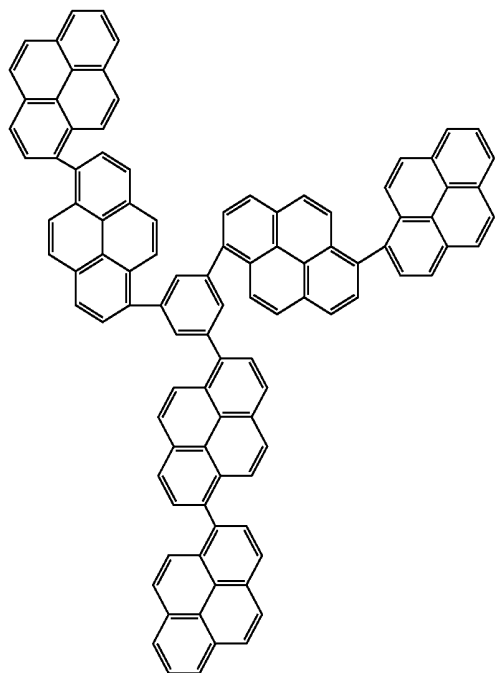


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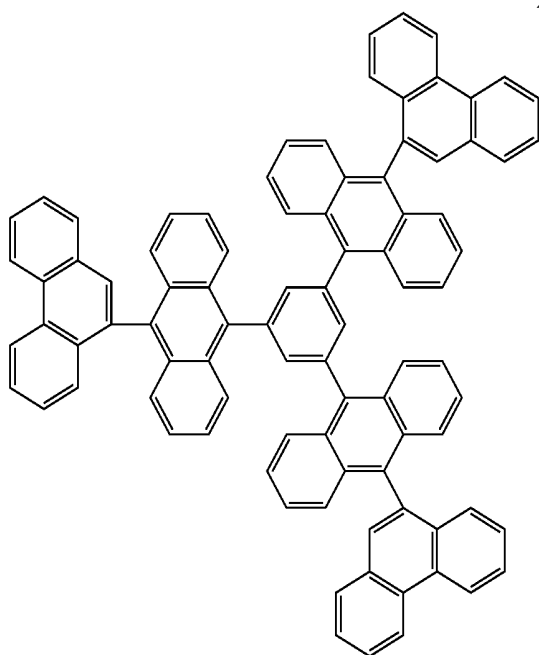
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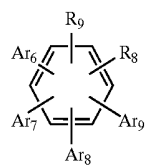
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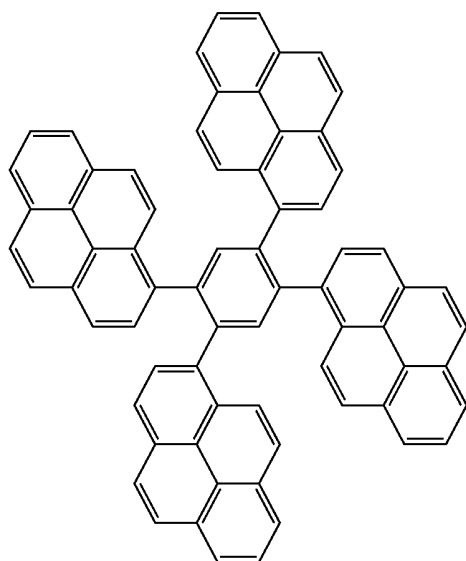


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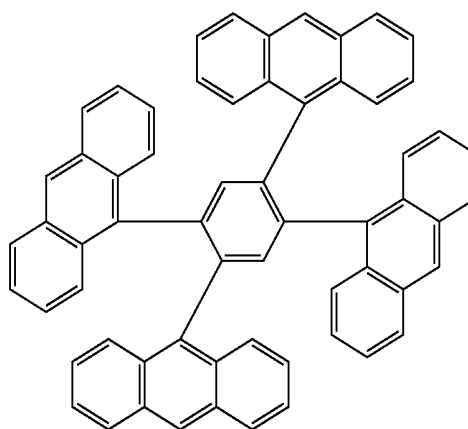


Formula [III]

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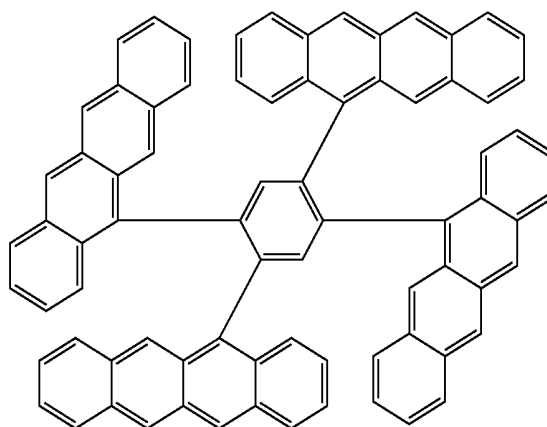
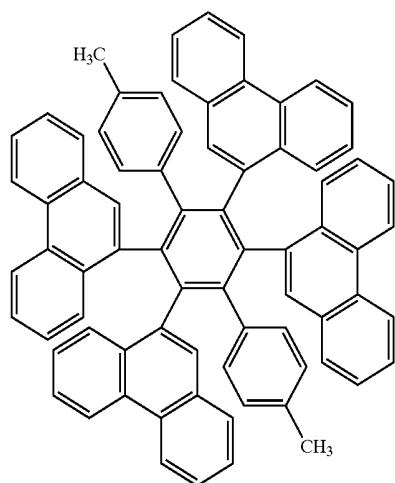
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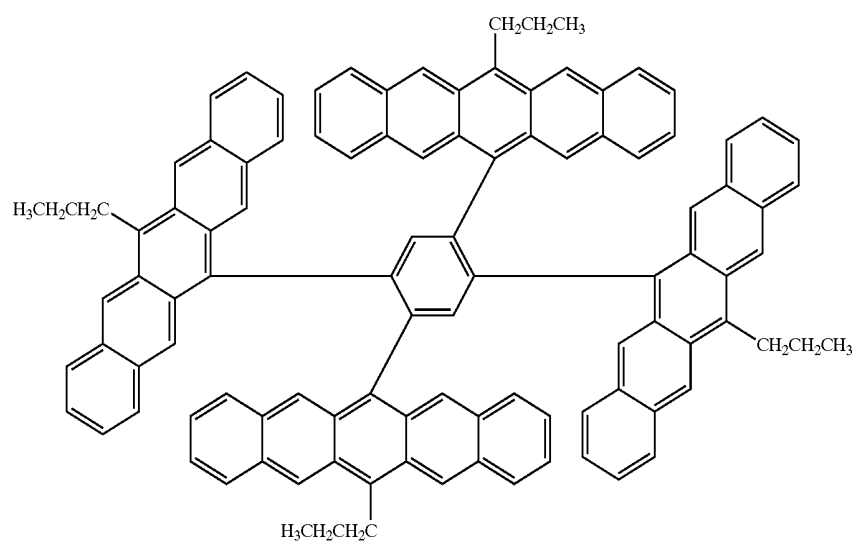
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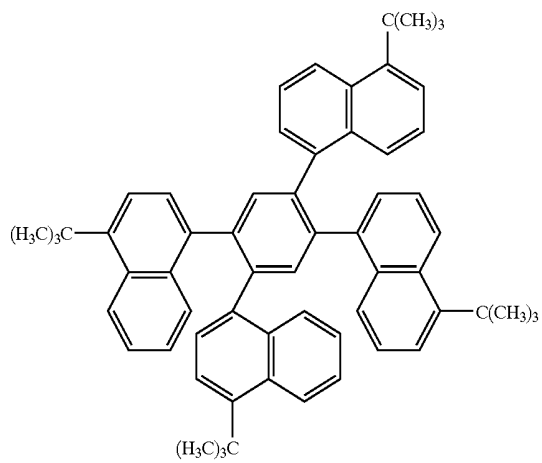
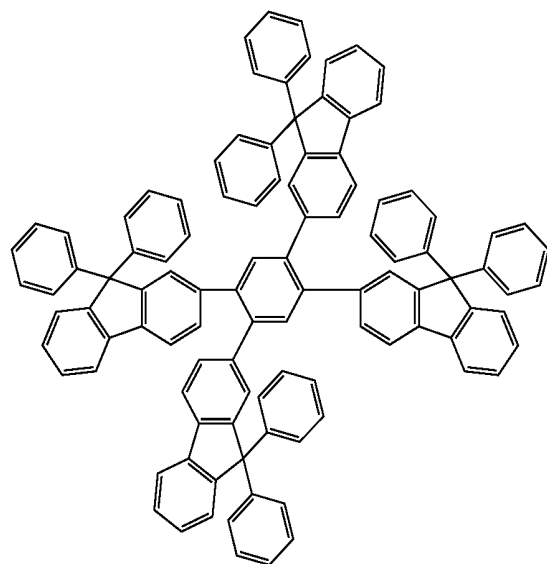


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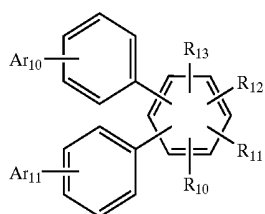
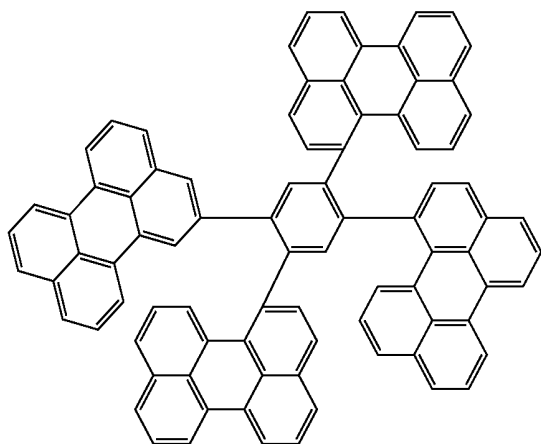


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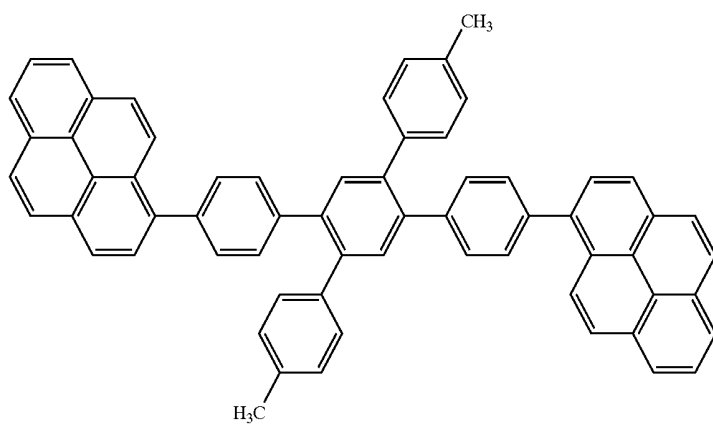
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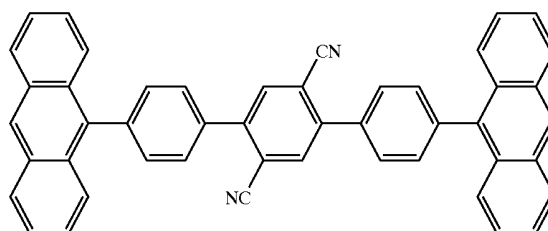
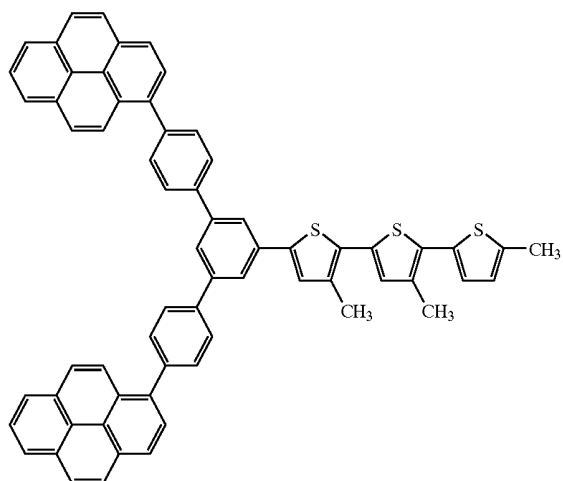
Formula [IV]

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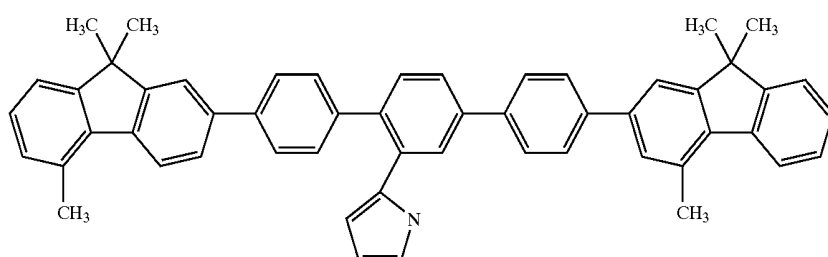
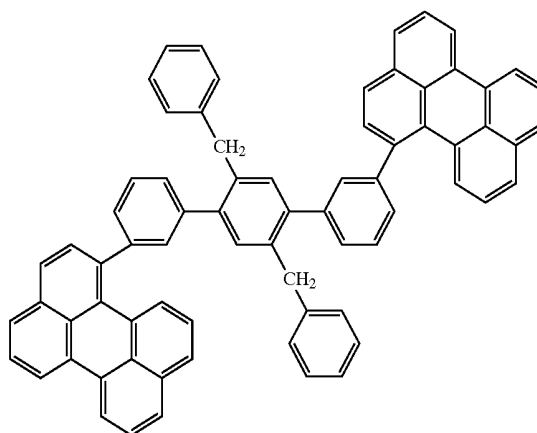
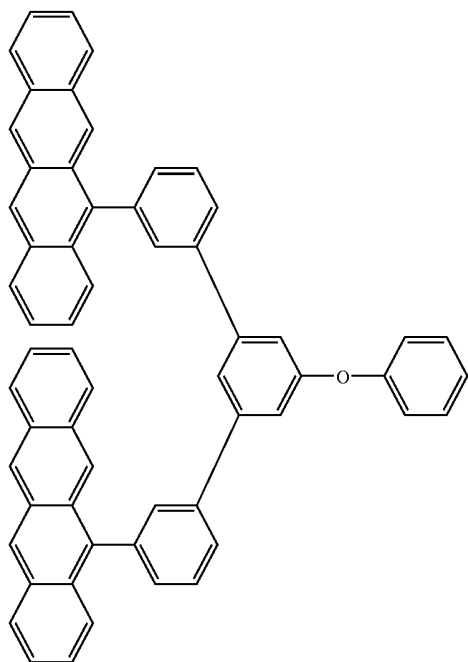
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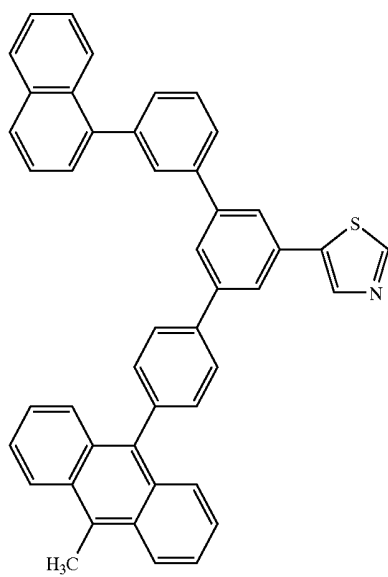
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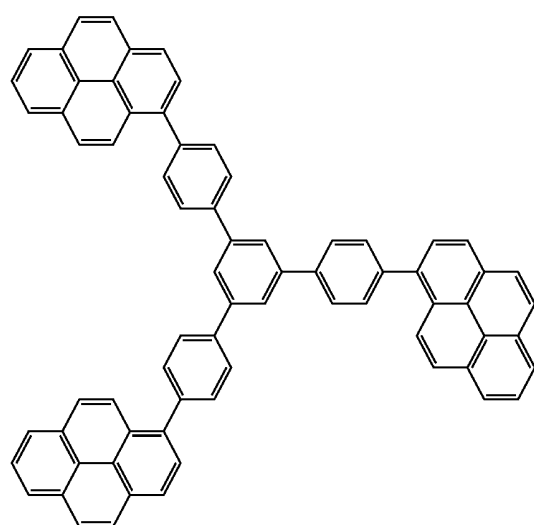
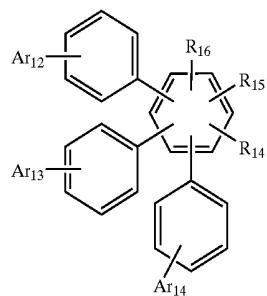
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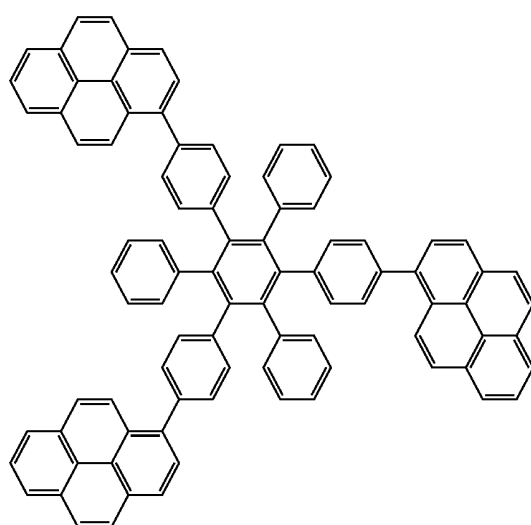
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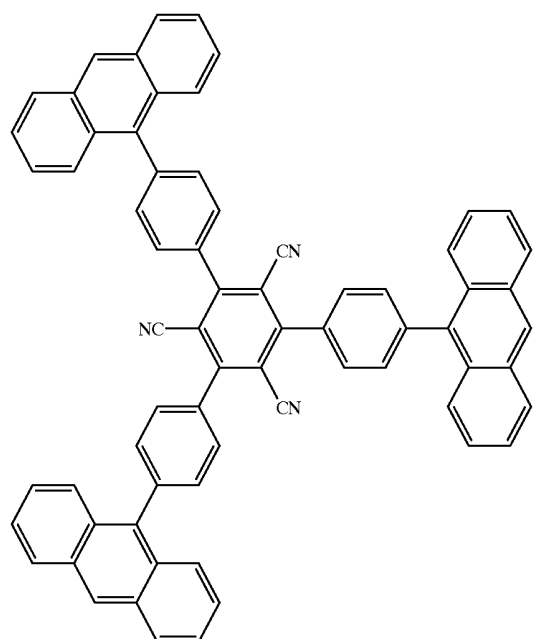
Formula [V]



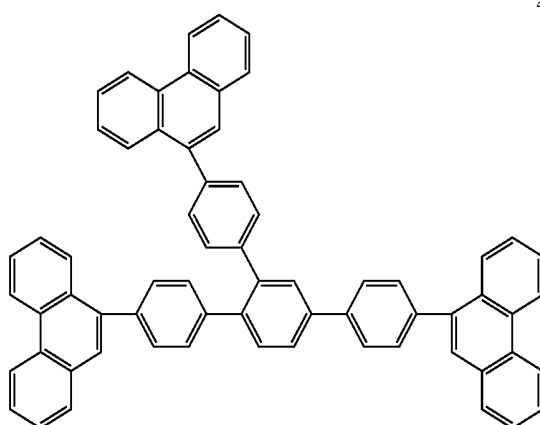
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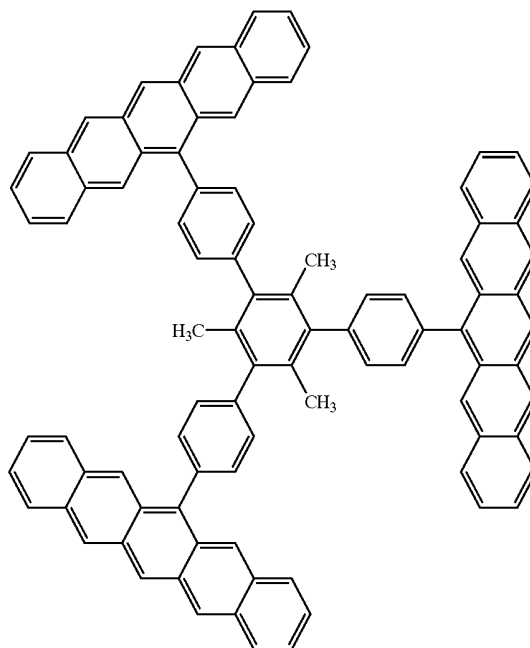
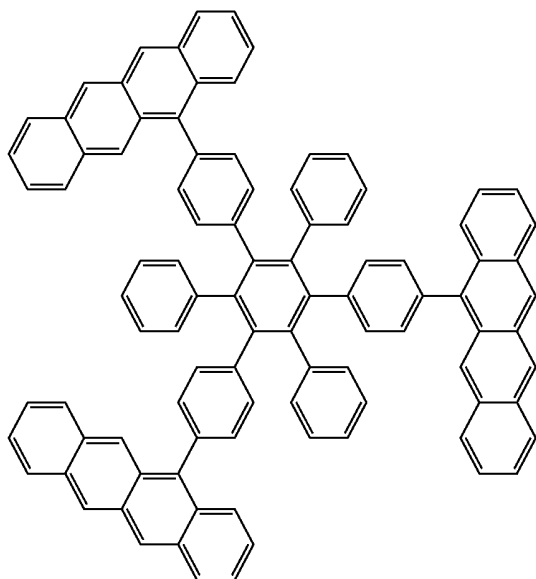
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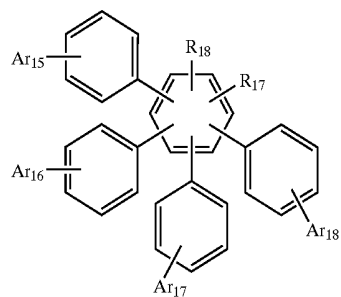
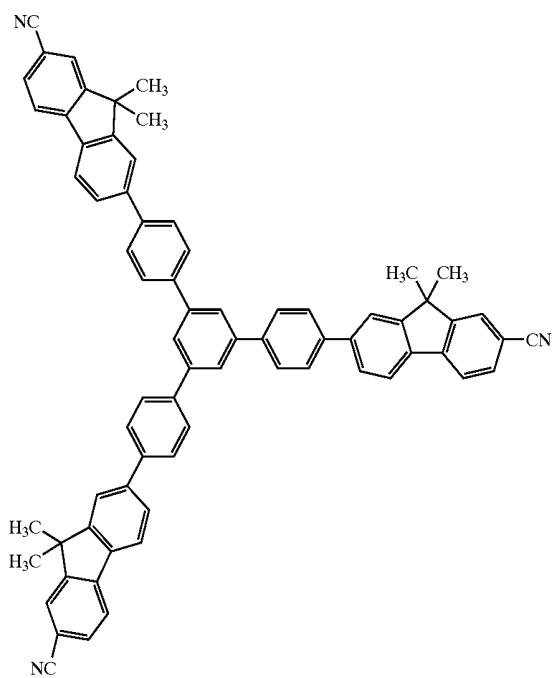
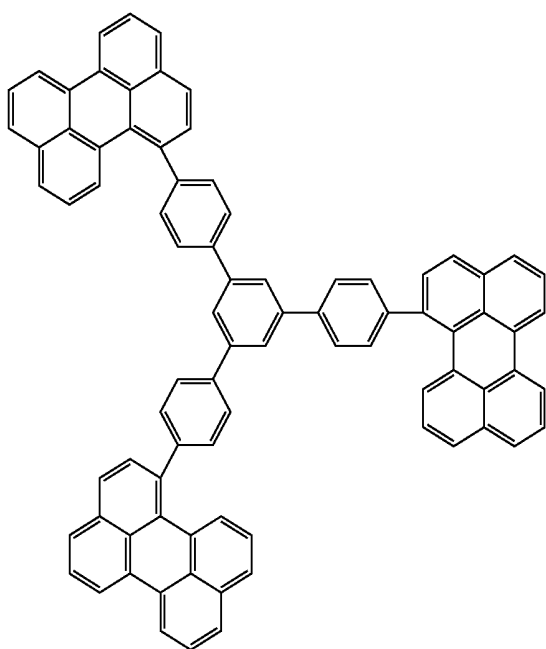
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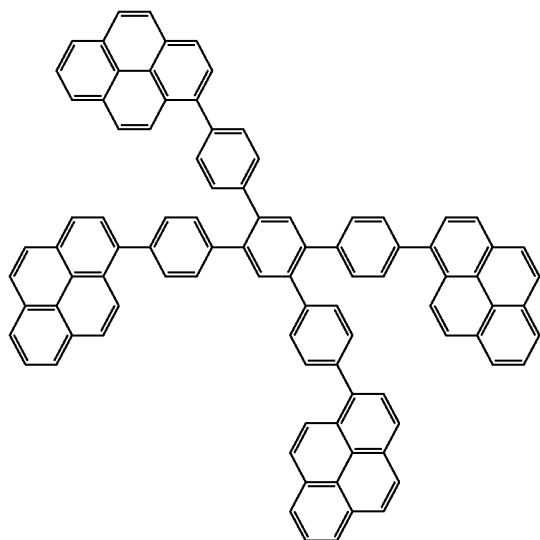
Formula [VI]

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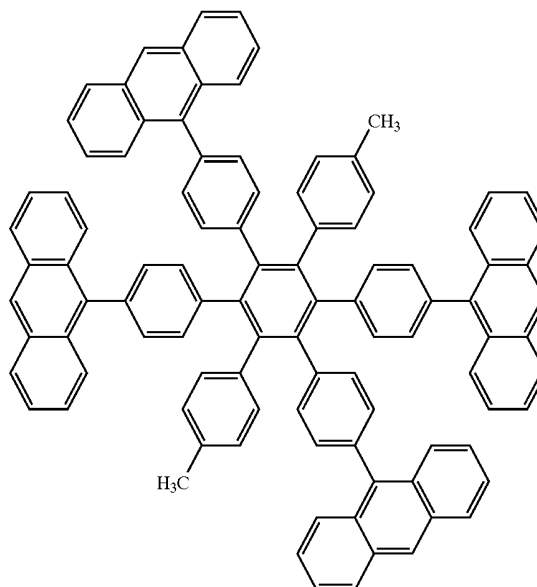
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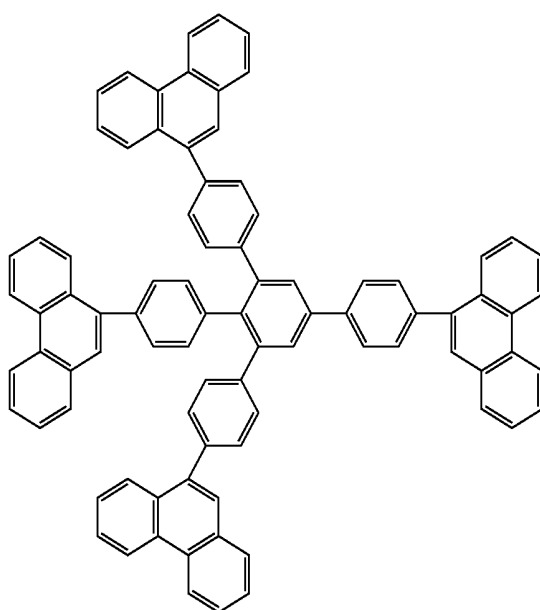
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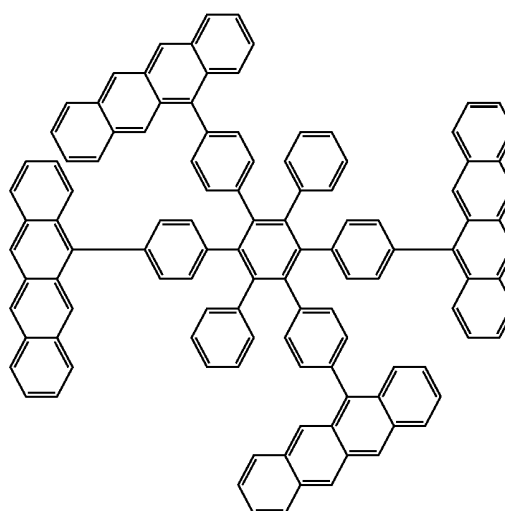
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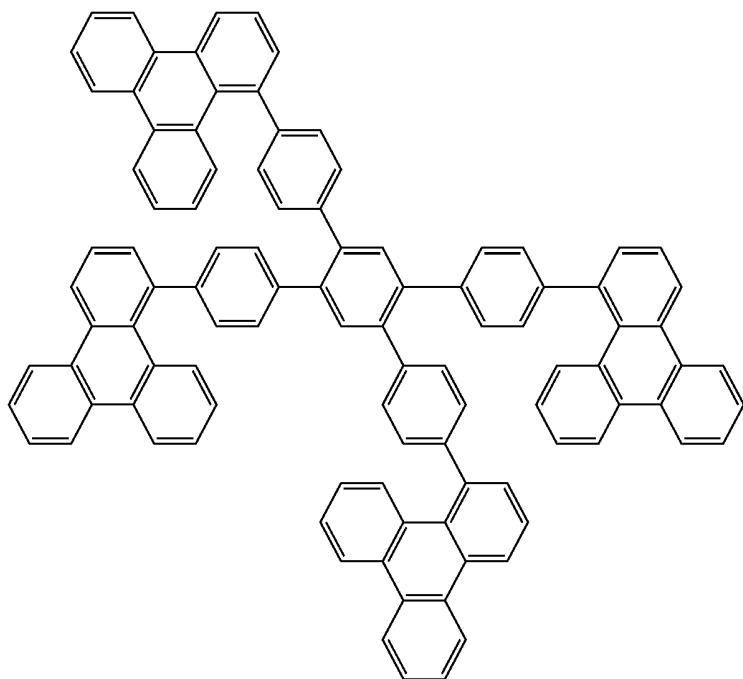


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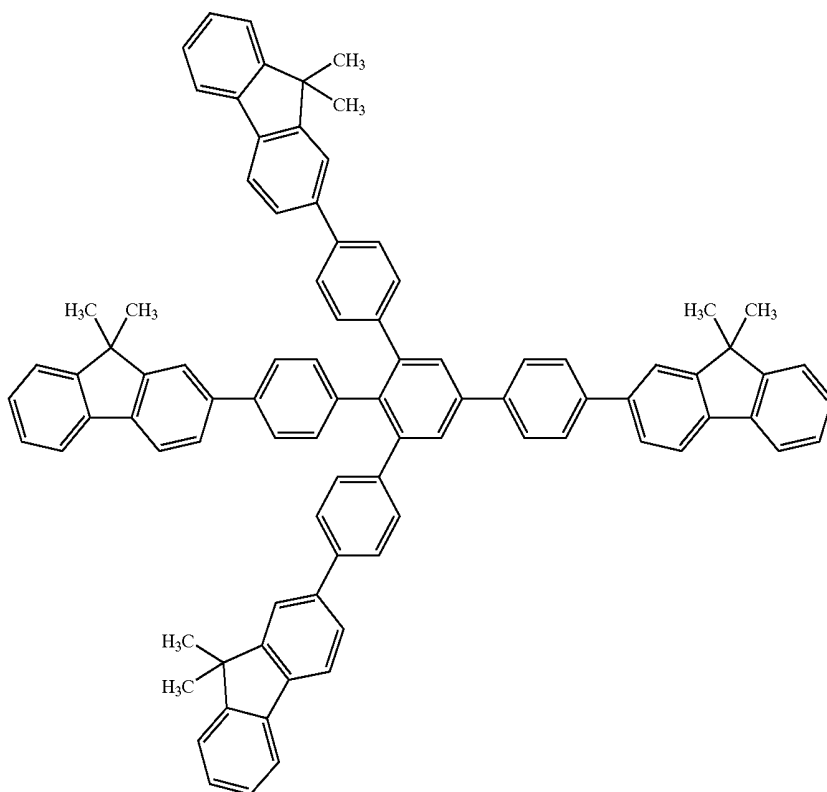
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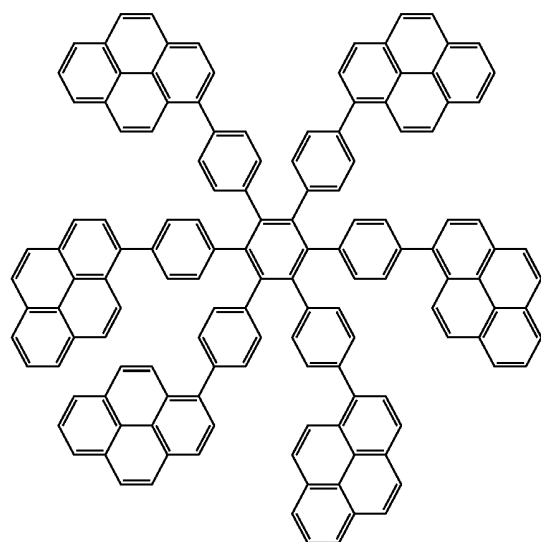
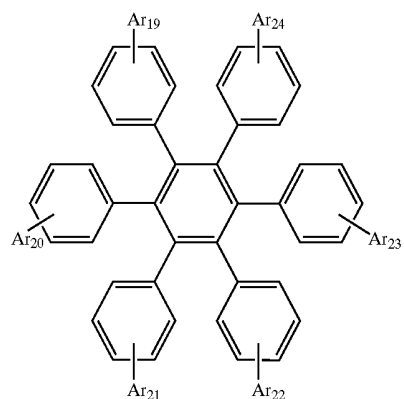


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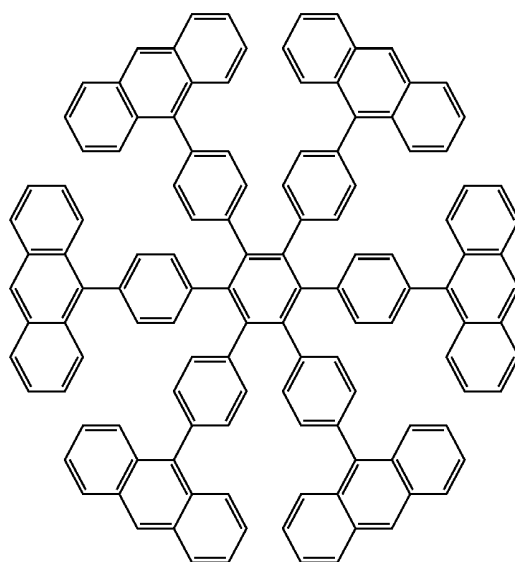
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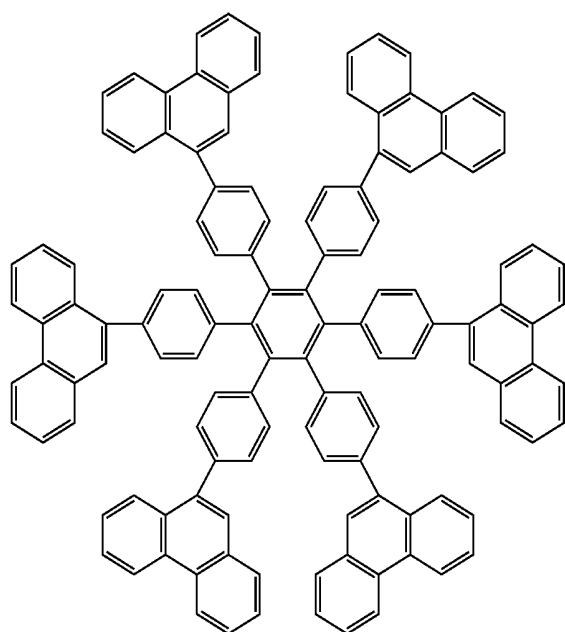
Formula [VII]



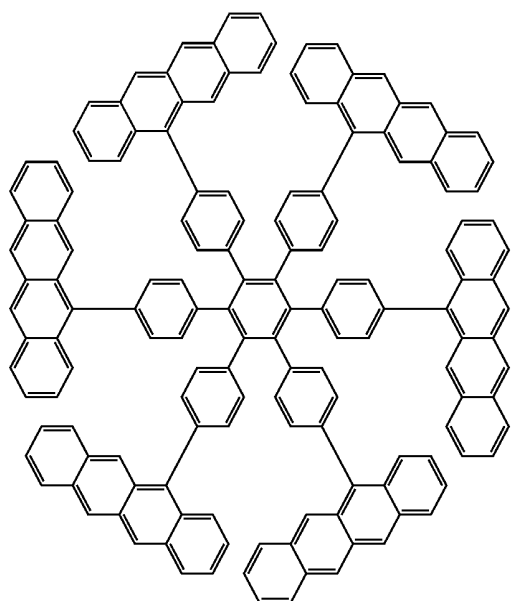
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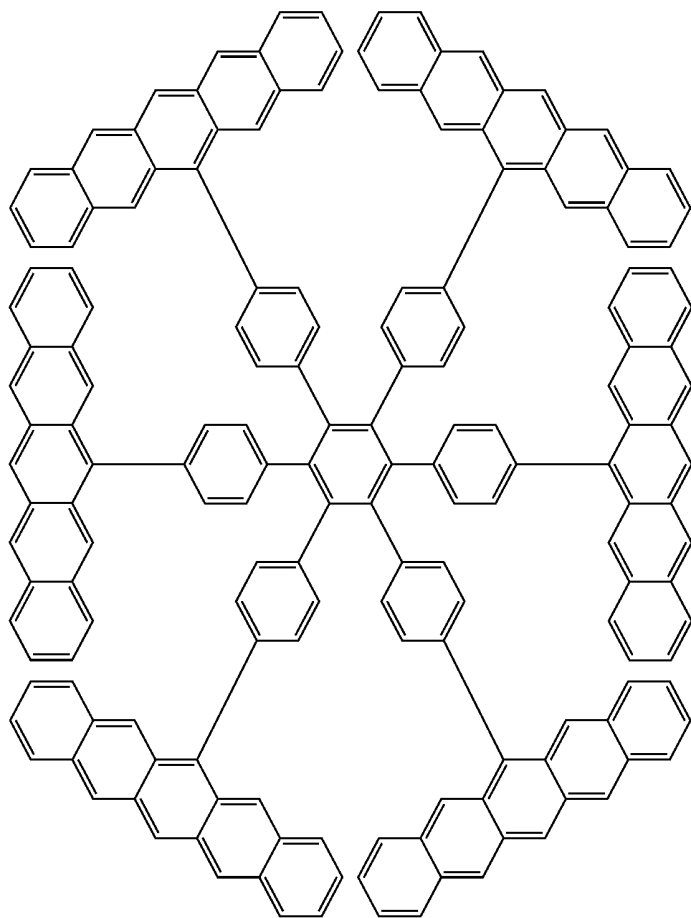
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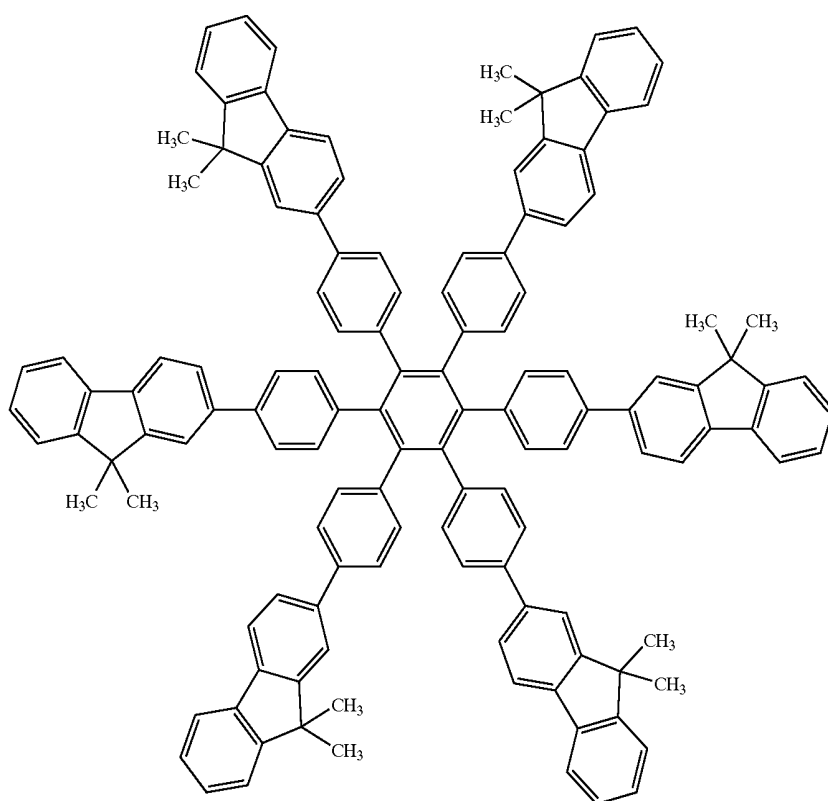
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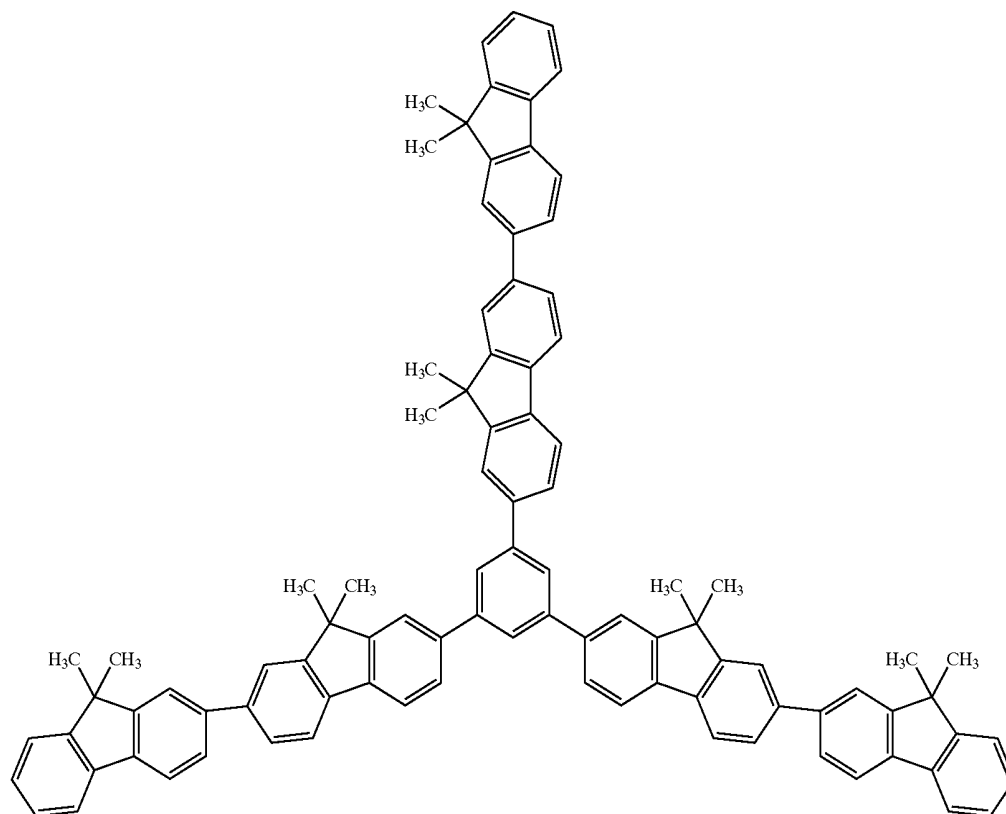
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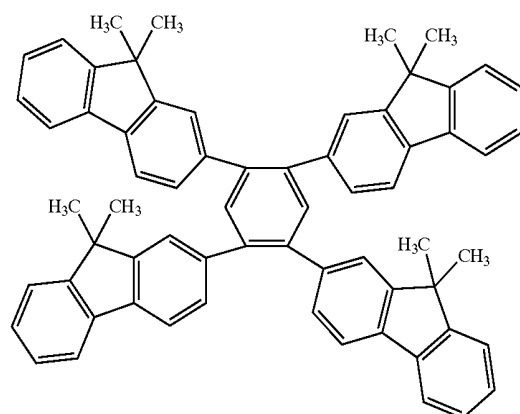
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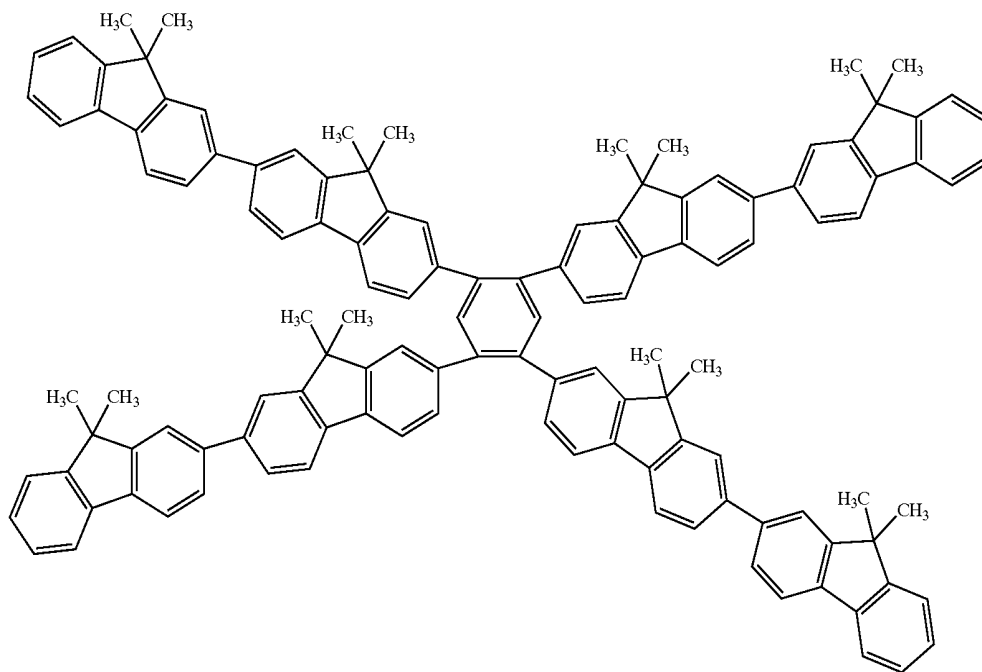
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The fused polynuclear compounds of the formulas (I) to (VII) used in the present invention may be synthesized through ordinary processes including Suzuki Coupling process using a palladium catalyst (e.g., "Chem. Rev.", 95, 2457-2483 (1995)), Yamamoto process using a nickel catalyst (e.g., "Bull. Chem. Soc. Jpn." 51, 2091 (1978)) and a process using an aryl tin compound (e.g., "J. Org. Chem.", 52, 3296 (1987)).

The fused polynuclear compound of the formulas (I) to (VII) used in the present invention is an excellent organic luminescence function material in terms of electron transfer performance, luminescence performance and durability, thus being a material suitable for an electron transport layer and/or a luminescence layer. Further when an organic luminescence function layer is formed by vacuum (vapor) deposition or wet coating using an appropriate solvent, the resultant organic luminescence function layer is less liable to cause crystallization, thus being excellent in stability with time.

In the present invention, as described above, the organic luminescence function layer disposed between the pair of electrodes (anode and cathode) may be formed in a plurality of organic compound layers including at least one layer thereof comprising the above-mentioned fused polynuclear compound of the formulas (I) to (VII). Further, such at least one layer may preferably function as an electron transport layer or a luminescence layer.

Each of the organic compound layers (organic luminescence function layers) may preferably be formed in a thickness of at most 10 μm , more preferably at most 0.5 μm , most preferably 0.01-0.5 μm .

Hereinbelow, layer structures of the organic luminescence device according to the present invention will be described specifically with reference to FIGS. 1 to 6 each illustrating an embodiment thereof.

Referring to FIGS. 1-6 the respective organic luminescence devices basically include a substrate 1, an anode 2 disposed on the substrate 1, one or more organic luminescence function layer disposed on the anode 2, and a cathode 4 disposed on the one or more organic luminescence function layer.

In an embodiment shown in FIG. 1, the organic luminescence function layer is composed of a single layer exhibiting multifunctional performances in terms of a hole transport ability, an electron transportability and a luminescence performance. These performances may be given by using a single multifunctional compound or by mixing the respective functional materials in a single layer.

In another embodiment shown in FIG. 2, the organic luminescence function layer is composed of a hole transport layer 5 disposed on the anode 2 and an electron transport layer 6 disposed on the hole transport layer 5. In this embodiment, a luminescent material also exhibits either one or both of a hole transport performance and an electron transport performance and is used in combination with a hole transport material free from a luminescence performance or an electron transport material free from a luminescence performance. In this embodiment, either one of the hole transport layer 5 and the electron transport layer 6 also functions as in the luminescence layer.

In another embodiment shown in FIG. 3, the organic luminescence function layer is composed of three layers consisting of a hole transport layer 5, a luminescence layer 3 and an electron transport layer 6 disposed in this order on the anode 2. In this embodiment, carrier (hole/electron) transport performances and luminescence performance are functionally separated into the three layers which may appropriately be formed by using respective functional materials exhibiting a hole transport performance, an electron transport performance and a luminescence performance.

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mance. As a result, it is possible to allow not only an increased latitude in selection of materials but also use of various compounds different in emission wavelength, thus resulting in a variety of emission hues. Further, it also becomes possible to effectively confining respective carriers or excitons in the luminescence layer **3** thus improving a luminescence-efficiency.

FIG. **4** shows another embodiment of the organic luminescence device of the present invention. Referring to FIG. **4**, the organic luminescence device has four layers as the organic luminescence function layers including three functional layers similar to those (hole transport layer **5**, luminescence layer **3** and electron transport layer **6**) shown in FIG. **3** and a hole injection layer **7** disposed between the anode **2** and the hole transport layer **5**. The use of the hole injection layer **7** is effective in improving adhesive properties between the anode **2** and the hole transport layer **5** or hole injection performance, thus resulting in luminescence at a low applied voltage.

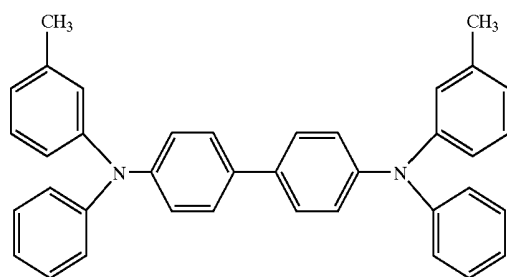
FIGS. **5** and **6** show other embodiments similar to those shown in FIGS. **3** and **4**, respectively, except that a hole (or exciton) blocking layer **8** for blocking passing of holes or excitons to the anode side is disposed between the electron transport layer **6** and the luminescence layer **3**. In these embodiments, by using a compound exhibiting a very high ionization potential in the hole blocking layer **8**, a resultant luminescence efficiency is effectively improved.

The layer structure of organic luminescence device of the present invention using the above-mentioned fused polynuclear compound is not restricted to those described above with reference to FIGS. **1-6** illustrating basic device structures of the organic luminescence device of the present invention.

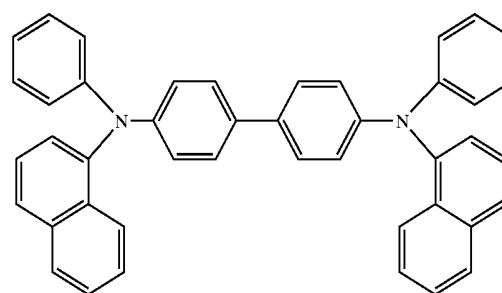
For example, the layer structure of the organic luminescence device according to the present invention may be modified by additionally forming an insulating layer, an adhesive layer or an interference layer at a boundary between an electrode and an organic luminescence function layer. Further, the hole transport layer **5** may be composed of two layers different in ionization potential.

The fused polynuclear compound used in the present invention (represented by any one of the formulas (I) to (VII)) may preferably be used as a material suitable for the electron transport layer and/or the luminescence layer but may be used in combination with known hole transport compounds, luminescent compounds and/or electron transport compounds shown below.

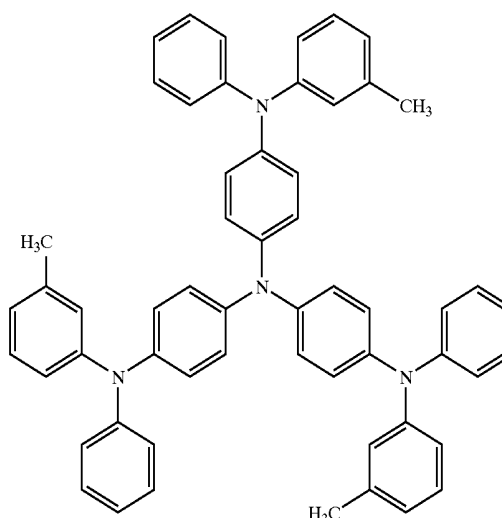
Hole Transport Material



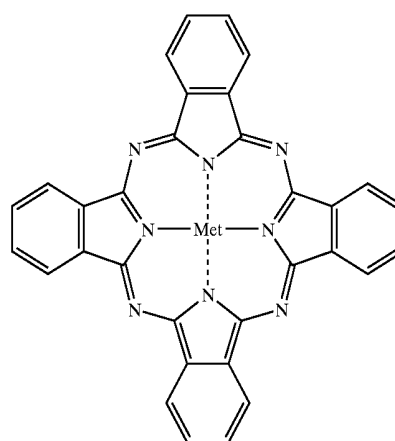
TPD



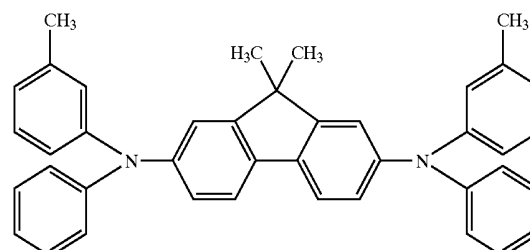
a-NPD



m-MTDATA



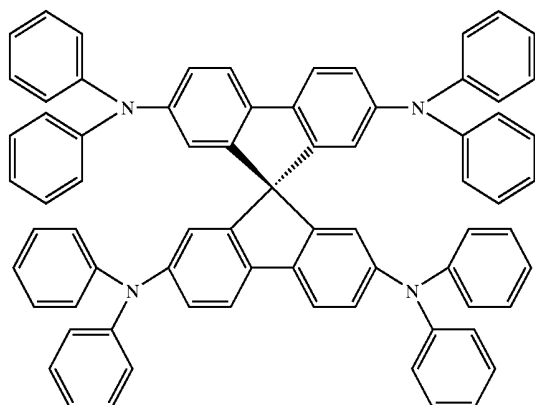
Met-Pc
Met = Cu, Mg, AlCl, TiO₂, SiCl₂, etc.



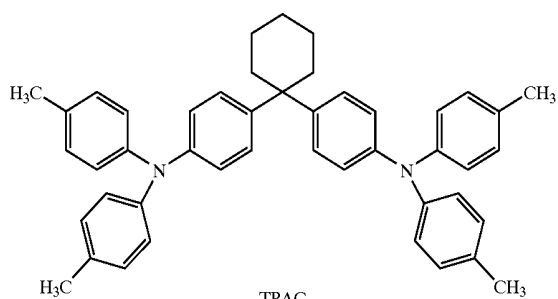
DTDPFL

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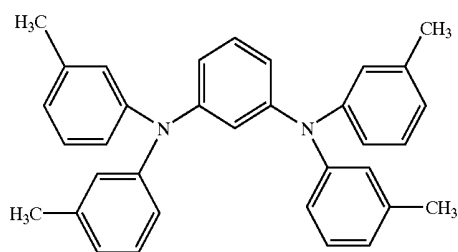
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spiro-TPD

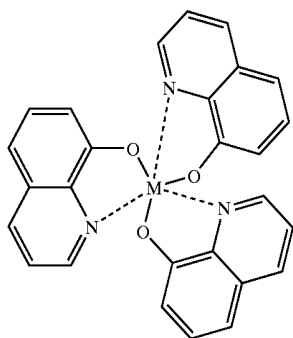


TPAC



PDA

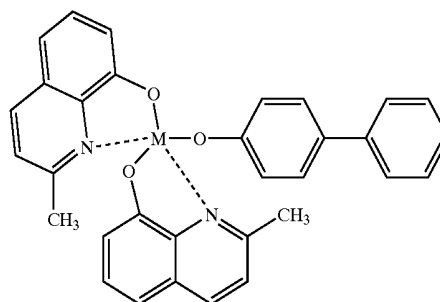
Electron Transport Luminescence Material



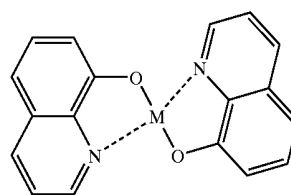
M = Al, Ga

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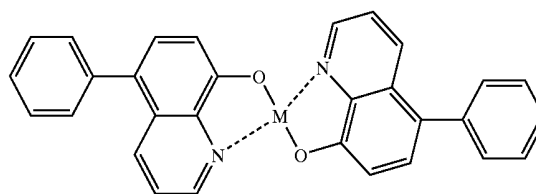
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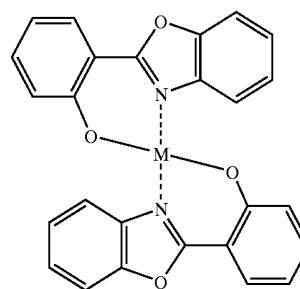
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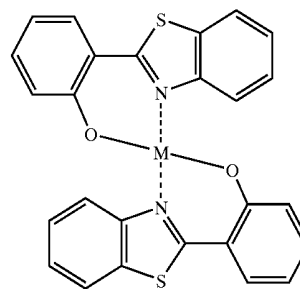
M = Zn, Mg, Be



M = Zn, Mg, Be



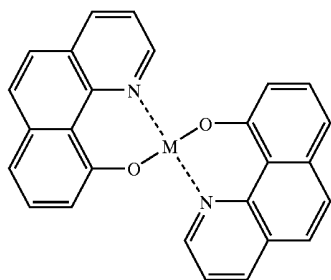
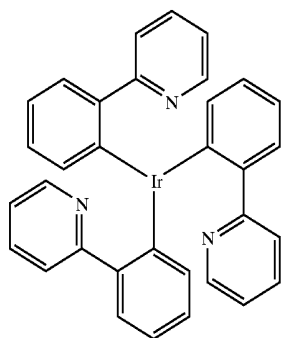
M = Zn, Mg, Be



M = Zn, Mg, Be

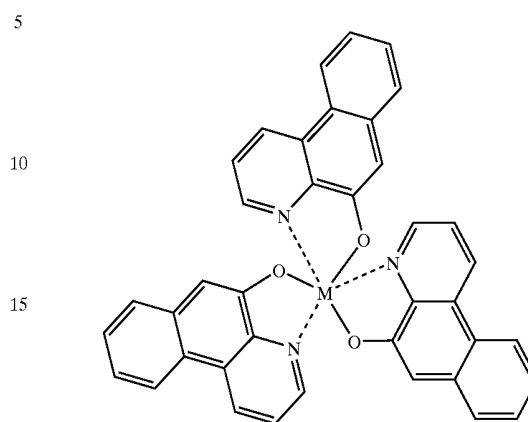
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$$M = \text{Zn, Mg, Be}$$

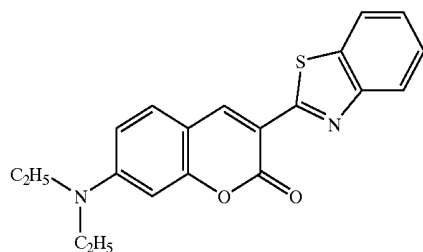
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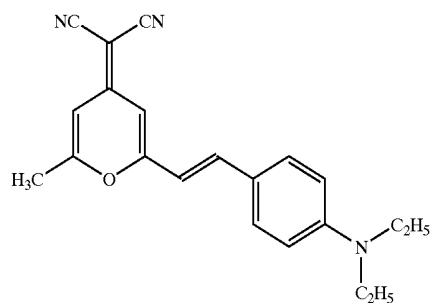


M = Al, Ga

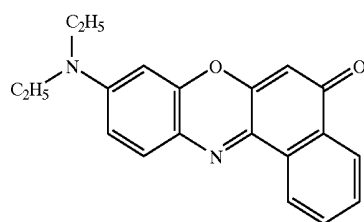
Luminescence Material



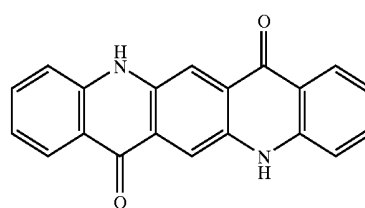
Coumarin 6



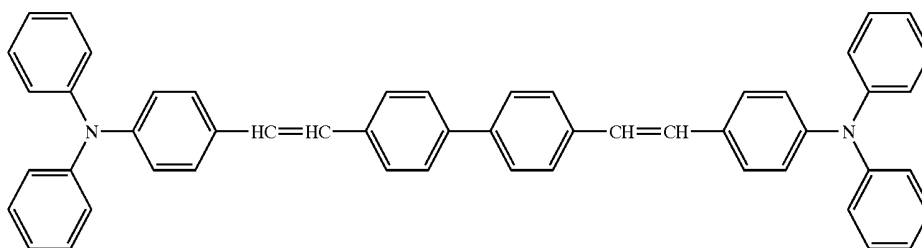
DCM-1



Nile red

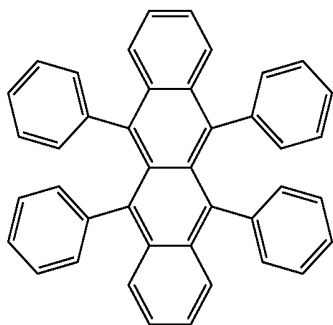


Quinacridone

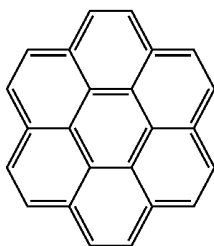


DPABVi

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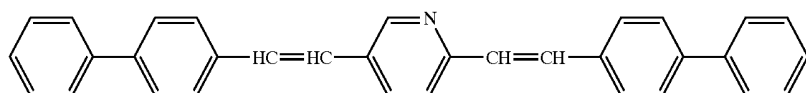
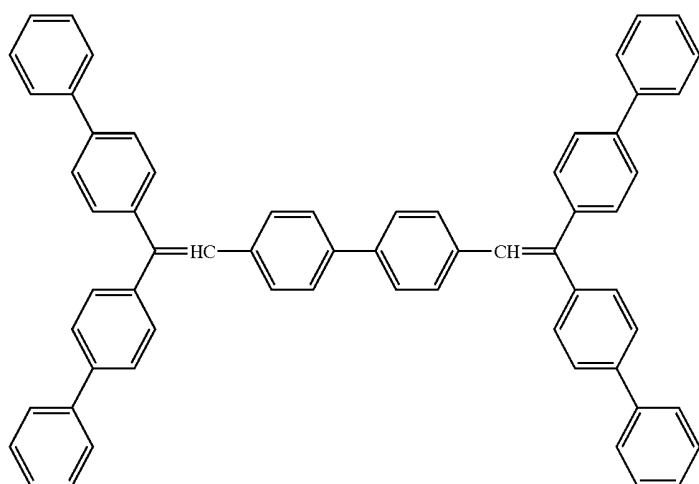
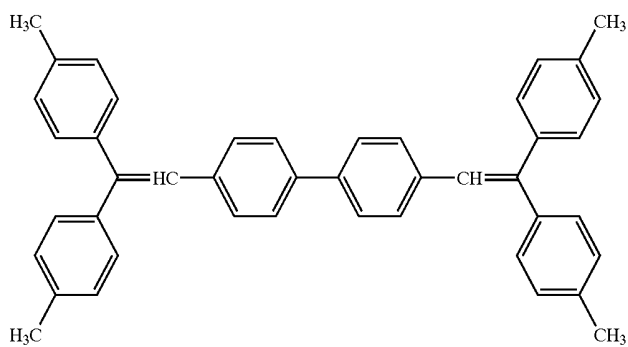


Rubrene



Coronene

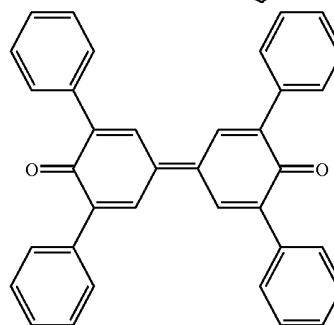
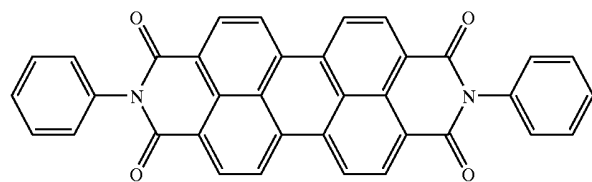
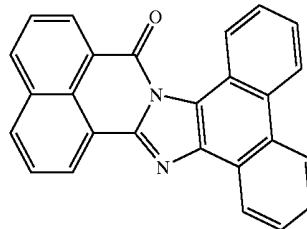
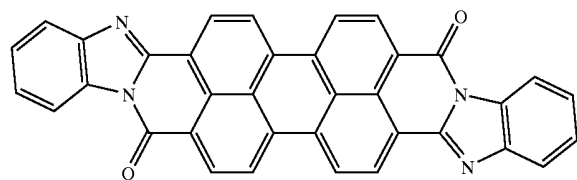
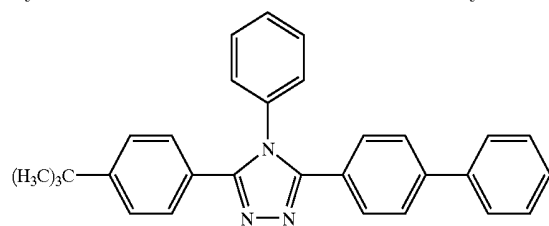
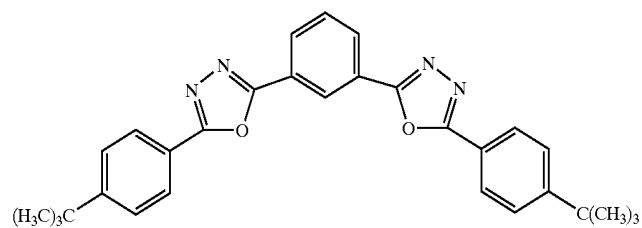
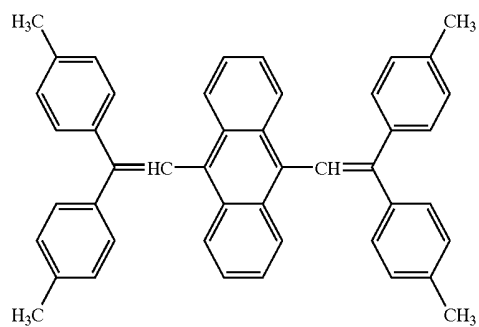
Luminescence Layer Matrix (Host) Material and Electron
Transport Material



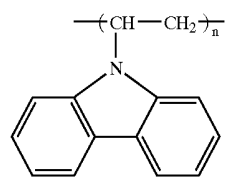
51

52

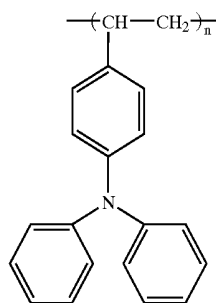
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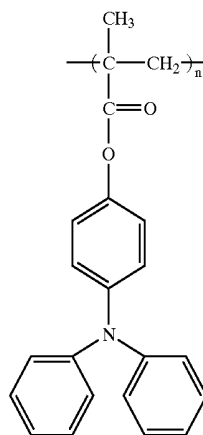
Polymer-type Hole Transport Material



PVCz



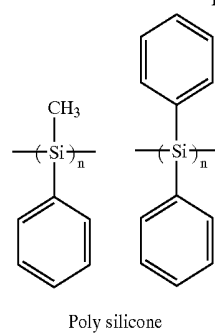
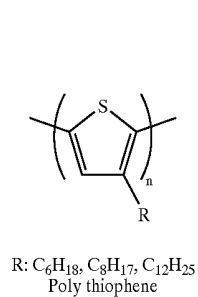
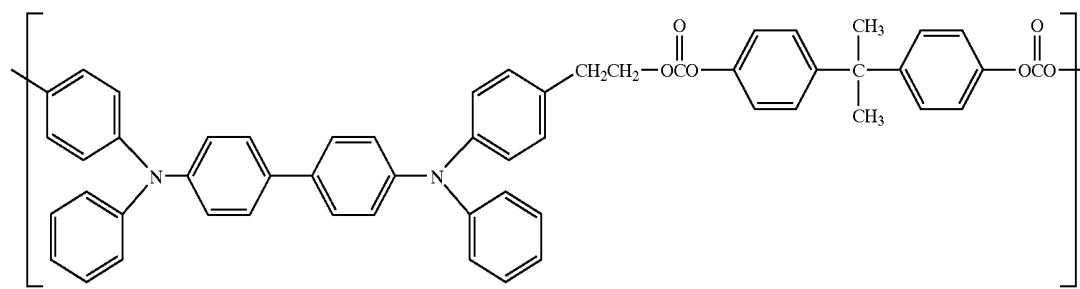
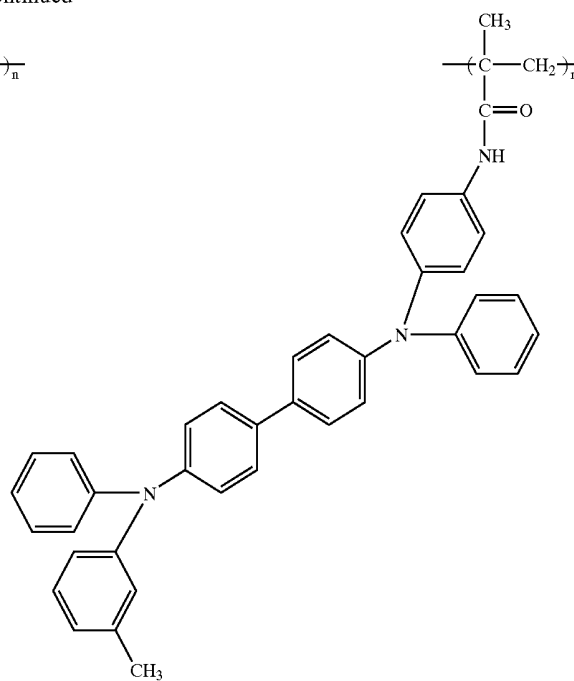
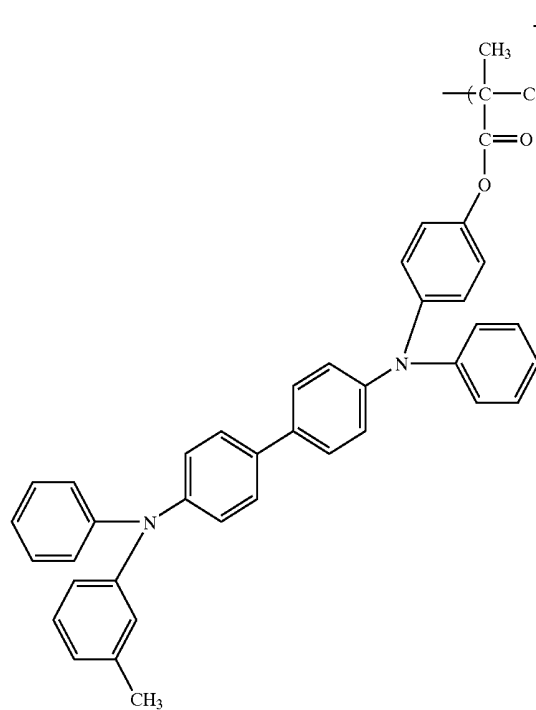
DPA-PS



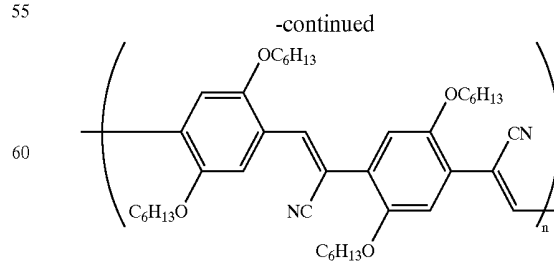
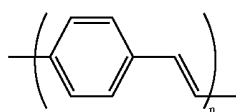
TPA-PMMA

53

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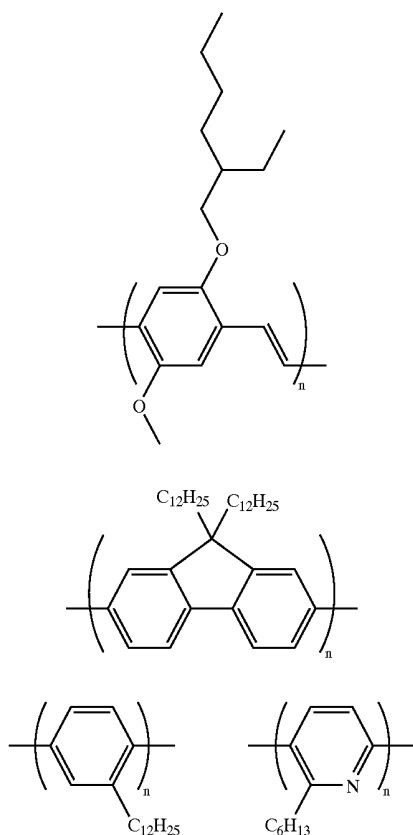


Polymer-type Luminescence Material and Charge Transport Material



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-continued



In the organic luminescence device according to the present invention, a layer of the fused polynuclear compound represented by the formulas (I) to (VII) and other layers comprising organic compounds may generally be formed in a thin film by vacuum deposition or wet coating using an appropriate solvent for dissolving such organic compounds (including the fused polynuclear compound). Particularly, in the case of using the wet coating, it is also possible to form a film in combination with an appropriate binder resin.

The binder resin may appropriately be selected from various known binder resins. Examples of the binder resin may include: polyvinyl carbazole, polycarbonate resin, polyester resin, polyarylate resin, polystyrene resin, acrylic resin, methacrylic resin, butyral resin, polyvinyl acetal resin, diallyl phthalate resin, phenolic resin, epoxy resin, silicone resin, polysulfone resin and urea resin. These resins may be used singly (as a homopolymer) or in combination of two or more species (as a copolymer).

The anode (electrode) constituting the organic luminescence device of the present invention may desirably be formed of a material having a work function as large as possible. Examples of such a material may include: metals such as gold, platinum, nickel, palladium, cobalt, selenium and vanadium; alloys of those metals; and metal oxides such as tin oxide, zinc oxide, indium tin oxide (ITO) and indium zinc oxide. Further, it is also possible to use electroconductive polymers such as polyaniline, polypyrrole, polythiophene, and polyphenylene sulfide. These materials may be used singly or in mixture.

On the other hand, the cathode (electrode) may desirably be formed of a material having a work function as small as

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possible. Examples of such a material may include: metals such as lithium, sodium, potassium, calcium, magnesium, aluminum, indium, silver, lead, tin and chromium; and alloys of these metals. It is also possible to use metal oxides such as ITO. The cathode may be formed in a single layer or plural layers.

The substrate for the organic luminescence device of the present invention is not particularly limited. Examples of the substrate may include an opaque substrate such as a metal substrate or ceramic substrate, and a transparent substrate such as glass substrate, quartz substrate or plastic sheet. Further, it is also possible to control emission light by using a color filter film, a fluorescent color conversion film or a dielectric reflection film, in combination with the substrate.

The organic luminescence device of the present invention may further comprise a protective layer or a sealing layer in order to prevent contact of the organic luminescence device with ambient oxygen or moisture.

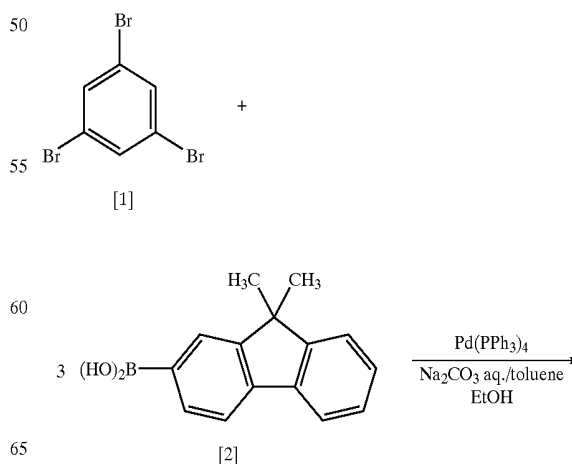
Examples of the protective film may include: a diamond film, a film of inorganic material such as metal oxide or metal nitride, a film of polymer such as fluorine-containing resin, polyparaxylene, polyethylene, silicone resin or polystyrene, and a photo-curable resin. Further, it is possible to effect packaging of the organic luminescence device per se by covering, e.g., glass substrate, gas-impermeable film or metal film with an appropriate sealing resin.

Incidentally, it is possible to cause white luminescence by incorporating a yellow luminescence material into the organic luminescence device of the present invention. For example, when rubrene as the yellow luminescence material is contained in a hole transport layer, white luminescence can be realized as specifically substantiated in Example 64 appearing hereinafter.

Hereinbelow, the present invention will be described more specifically based on Examples but is not restricted to the Examples.

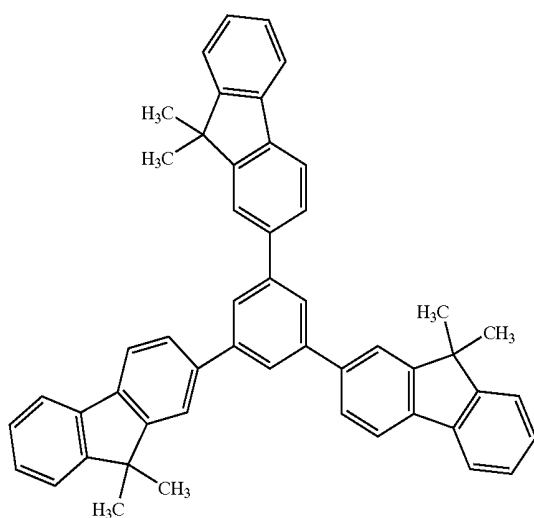
SYNTHESIS EXAMPLE 1

Synthesis of Ex. Comp. No. 22



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-continued



No. 22

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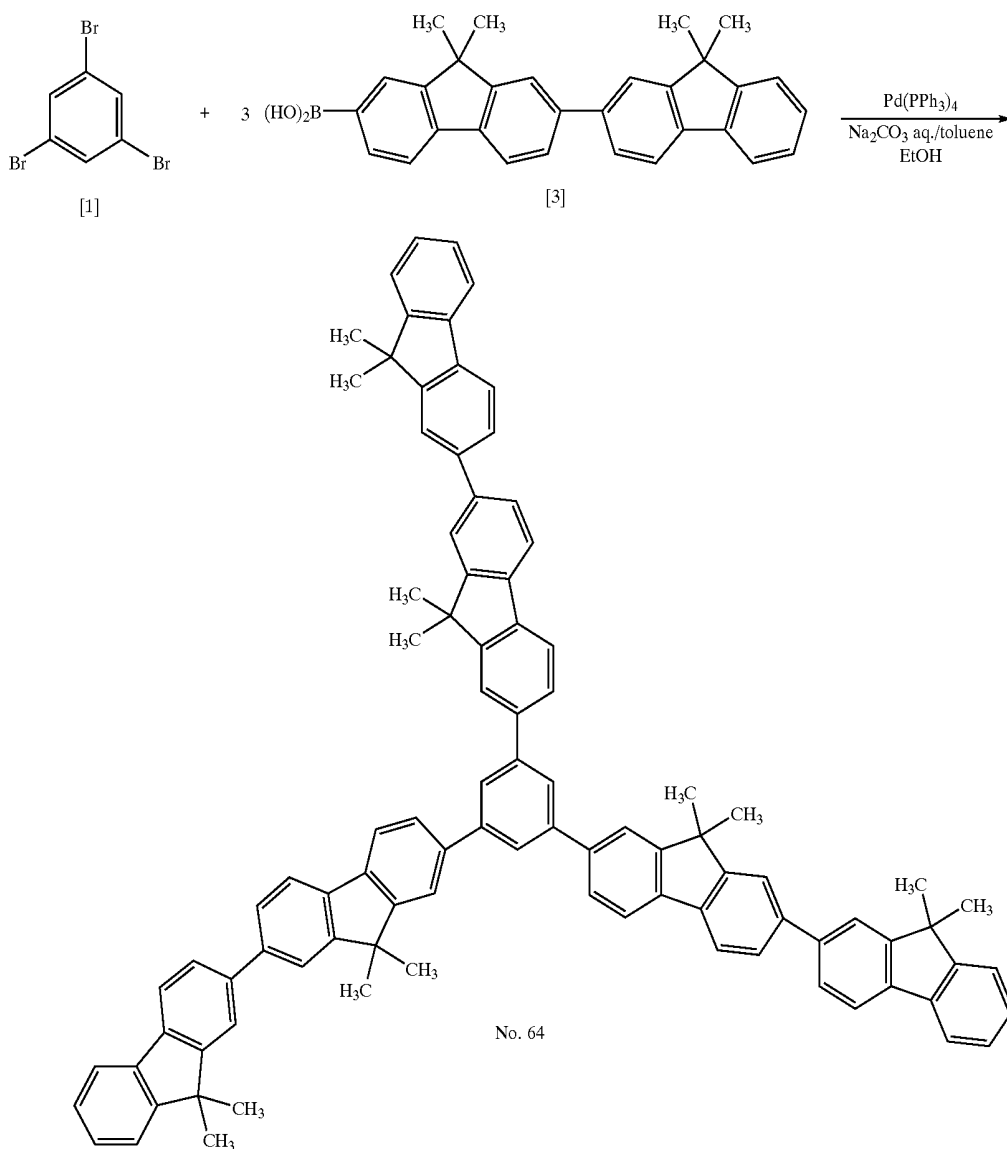
In a 500 ml-three necked flask, 0.8 g (2.52 mM) of 1,3,5-tribromobenzene[1], 3.0 g (12.6 mM) of fluorene-based boric acid [2], 160 ml of toluene and 80 ml of ethanol were placed and stirred in a nitrogen atmosphere at room temperature.

To the mixture, an aqueous solution of 15 g of sodium carbonate in 75 g of water was added dropwise and then 0.44 g (0.378 mM) of tetrakis (triphenylphosphine)palladium (0) was added. The system was stirred for 30 minutes at room temperature and heated up to 77° C., followed by stirring for 3 hours.

After the reaction, the reaction mixture was subjected to extraction of the organic layer with chloroform and dried with anhydrous sodium sulfate, followed by purification by silica gel column chromatography (eluent: hexane/toluene) to obtain 1.27 g of an objective compound (Ex. Comp. No. 22) (white crystal: Yield: 77%).

SYNTHESIS EXAMPLE 2

Synthesis of Ex. Comp. No. 64



No. 64

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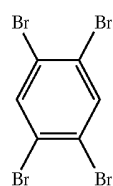
In a 500 ml-three necked flask, 0.8 g (2.52 mM) of 1,3,5-tribromobenzene[1], 4.8 g (12.6 mM) of fluorene-based boric acid [3], 160 ml of toluene and 80 ml of ethanol were placed and stirred in a nitrogen atmosphere at room temperature.

To the mixture, an aqueous solution of 15 g of sodium carbonate in 75 g of water was added dropwise and then 0.44 g (0.378 mM) of tetrakis (triphenylphosphine)palladium (0) was added. The system was stirred for 30 minutes at room temperature and heated up to 77° C., followed by stirring for 3 hours.

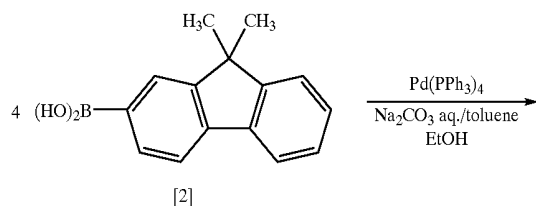
After the reaction, the reaction mixture was subjected to extraction of the organic layer with chloroform and dried with anhydrous sodium sulfate, followed by purification by silica gel column chromatography (eluent: hexane/toluene) to obtain 2.00 g of an objective compound (Ex. Comp. No. 64) (white crystal: Yield: 73%).

SYNTHESIS EXAMPLE 3

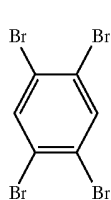
Synthesis of Ex. Comp. No. 65



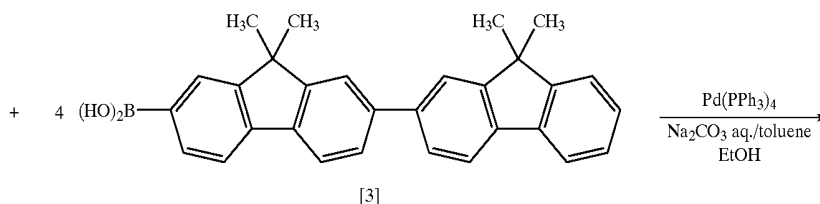
[4]



[2]



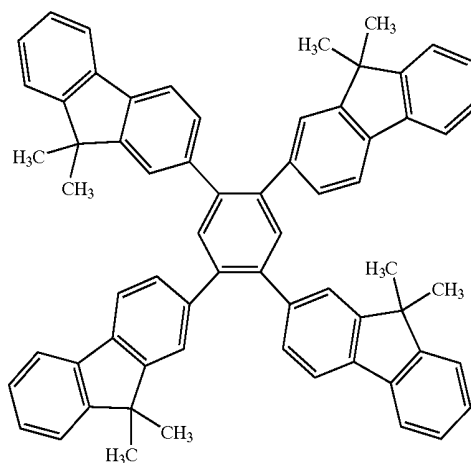
[4]



[3]

60

-continued



No. 65

In a 500 ml-three necked flask, 0.75 g (1.88 mM) of 1,2,4,5-tetrabromobenzene [4], 3.0 g (12.6 mM) of fluorene-based boric acid [2], 160 ml of toluene and 80 ml of ethanol were placed and stirred in a nitrogen atmosphere at room temperature.

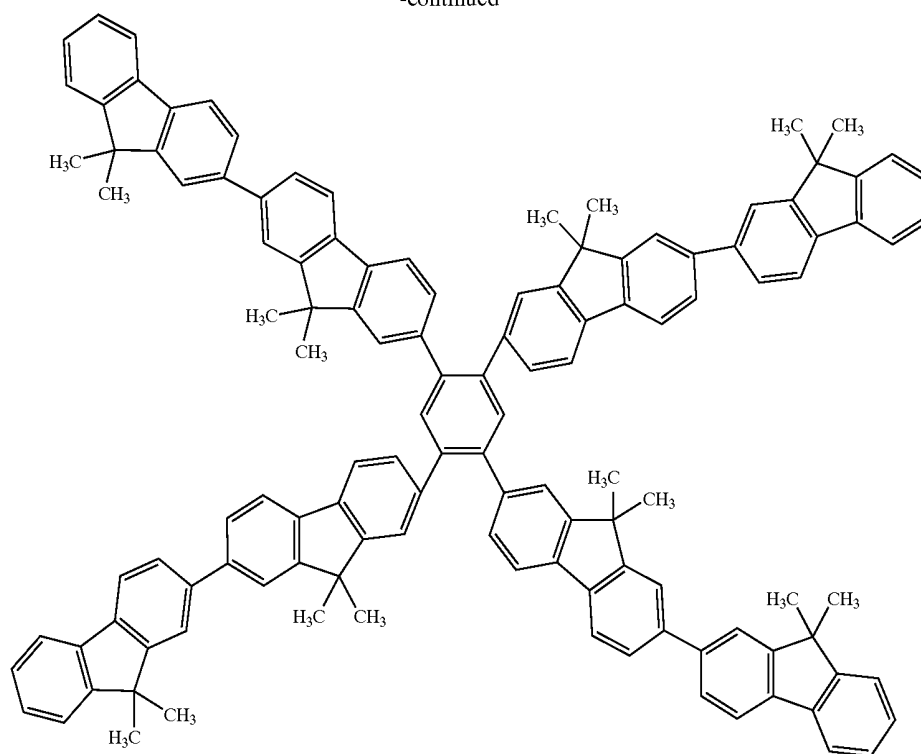
To the mixture, an aqueous solution of 15 g of sodium carbonate in 75 g of water was added dropwise and then 0.43 g (0.376 mM) of tetrakis (triphenylphosphine)palladium (0) was added. The system was stirred for 30 minutes at room temperature and heated up to 77° C., followed by stirring for 5 hours.

After the reaction, the reaction mixture was subjected to extraction of the organic layer with chloroform and dried with anhydrous sodium sulfate, followed by purification by silica gel column chromatography (eluent: hexane/toluene) to obtain 1.41 g of an objective compound (Ex. Comp. No. 65) (white crystal: Yield: 88%).

SYNTHESIS EXAMPLE 4

Synthesis of Ex. Comp. No. 66

-continued



No. 66

In a 500 ml-three necked flask, 0.75 g (2.52 mM) of 1,2,4,5-tetrabromobenzene [4], 4.8 g (12.6 mM) of fluorene-based boric acid [3], 160 ml of toluene and 80 ml of ethanol were placed and stirred in a nitrogen atmosphere at room temperature.

To the mixture, an aqueous solution of 15 g of sodium carbonate in 75 g of water was added dropwise and then 0.43 g (0.376 mM) of tetrakis (triphenylphosphine)palladium (0) was added. The system was stirred for 30 minutes at room temperature and heated up to 77° C., followed by stirring for 5 hours.

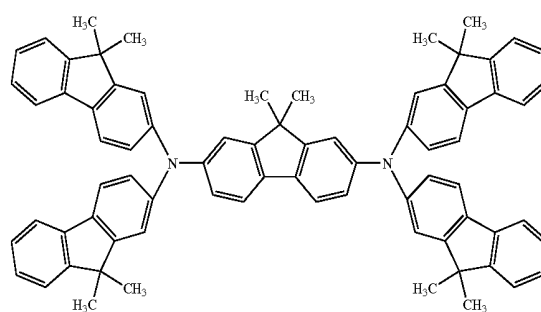
After the reaction, the reaction mixture was subjected to extraction of the organic layer with chloroform and dried with anhydrous sodium sulfate, followed by purification by silica gel column chromatography (eluent: hexane/toluene) to obtain 1.88 g of an objective compound (Ax. Comp. No. 66) (white crystal: Yield: 70%).

EXAMPLE 1

An organic luminescence device shown in FIG. 2 was prepared in the following manner.

On a 0.7 mm-thick glass substrate 1, a 120 nm-thick ITO (indium tin oxide) film (anode 2) was formed by sputtering to prepare a transparent electroconductive support, which was then successively subjected to ultrasonic cleaning with acetone and with isopropyl alcohol (IPA). The resultant transparent electroconductive support was then subjected to boiling leaning with IPA and was dried, followed by UV/ozone cleaning.

On the transparent electroconductive support, a solution of a hole transport material shown below in chloroform was applied by spin coating to form a 30 nm-thick hole transport layer 5.



On the hole transport layer 5, a 50 nm-thick electron transport layer 6 of a fused polynuclear compound (Ex. Comp. No. 1) was formed by vacuum deposition under conditions including a vacuum degree (pressure) of 1.0×10^{-4} Pa and a deposition rate of 0.2–0.3 nm/sec.

Then, on the electron transport layer 6, a 150 nm-thick metal electrode (cathode 4) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa/1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 10 volts was applied between the ITO electrode (anode 2, positive pole) and the Al—Li electrode (cathode 4, negative pole), whereby a current was passed through the

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organic luminescence device at a current density of 9.0 mA/cm² and blue luminescence was observed at a luminance of 750 cd/m².

Then, when the organic luminescence device was supplied with a voltage for 100 hours while keeping a current density of 7.0 mA/cm² in a nitrogen atmosphere, a luminance of 550 cd/m² (as initial luminance) was merely decreased to 470 cd/m² even after 100 hours of the voltage application, thus exhibiting a good durability. The results are also shown in Table 1 appearing hereinafter.

EXAMPLES 2-15

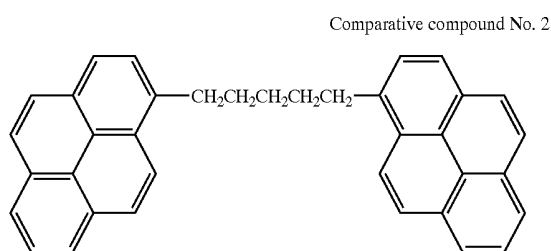
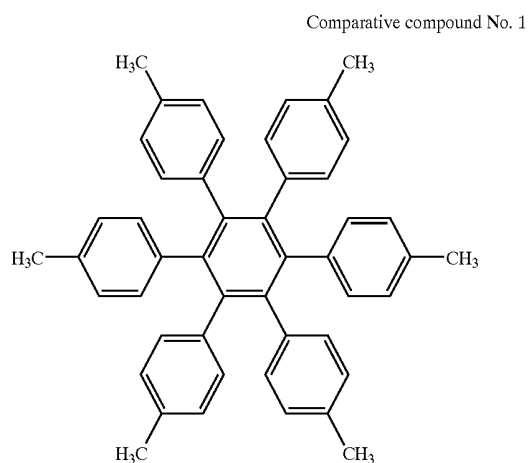
Organic luminescence devices were prepared and evaluated in the same manner as in Example 1 except that the fused polynuclear compound (Ex. Corp. No. 1) was changed to those (Ex. Comp. Nos. 5, 10, 16, 19, 24, 25, 31, 36, 39, 43, 46, 51, 58 and 63), respectively.

The results are shown in Table 1.

COMPARATIVE EXAMPLES 1-8

Organic luminescence devices were prepared and evaluated in the same manner as in Example 1 except that the fused polynuclear compound (Ex. Comp. No. 1) was changed to the following comparative compounds Nos. 1-8, respectively.

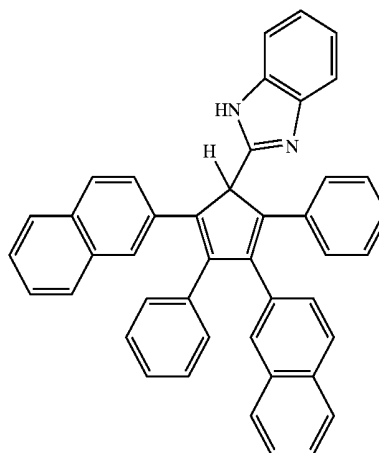
The results are shown in Table 2.



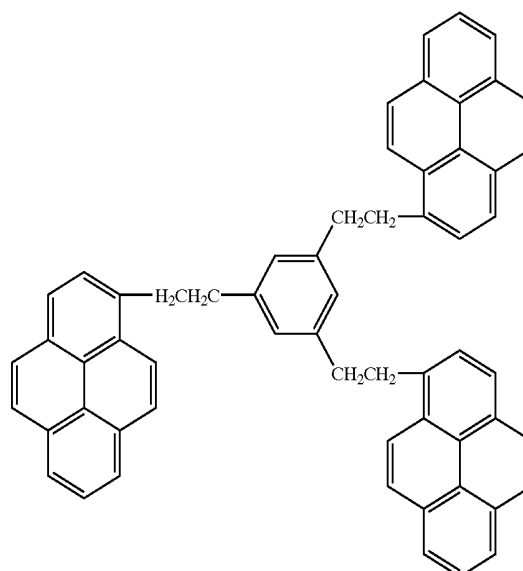
64

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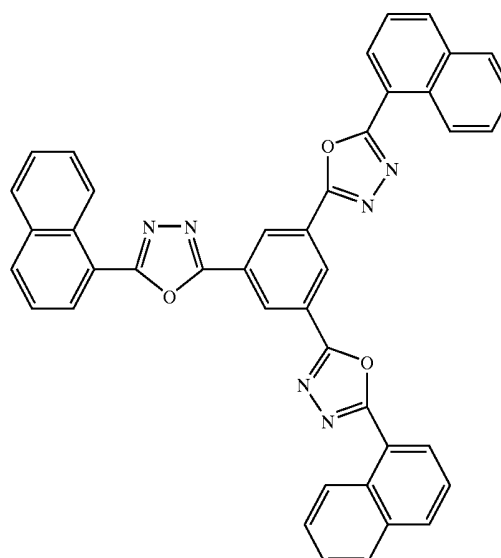
Comparative compound No. 3



Comparative compound No. 4

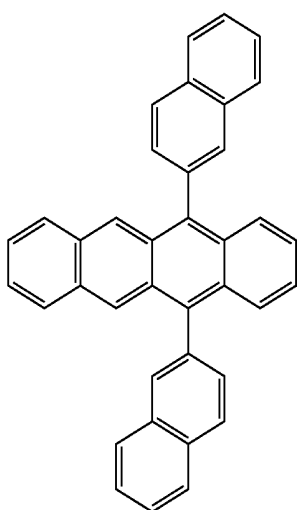


Comparative compound No. 5



65

-continued



Comparative compound No. 6

5

66

TABLE 1

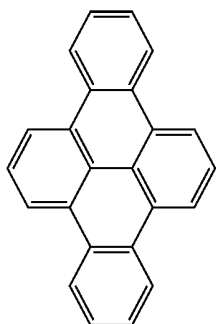
Initial					
Ex.		Applied	Luminance (at 7.0 mA/cm ²)		
Ex. No.	Comp. No.	voltage (V)	Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
1	1	10	750	550	470
2	5	10	640	480	440
3	10	10	670	510	470
4	16	10	1220	1130	810
5	19	10	880	690	610
6	24	10	560	530	430
7	25	10	920	780	650
8	31	10	960	830	740
9	36	10	580	490	430
10	39	10	660	520	460
11	43	10	570	510	440
12	46	10	720	570	510
13	51	10	710	550	490
14	58	10	1100	980	800
15	63	10	650	520	450

TABLE 2

Comp.		Initial			
Comp.	Ex.	Applied		Luminance (at 7.0 mA/cm ²)	
Ex. No.	Comp. No.	voltage (V)	Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
1	Comp. 1	10	140	100	10
2	Comp. 2	10	70	60	No luminescence
3	Comp. 3	10	90	70	No luminescence
4	Comp. 4	10	80	70	No luminescence
5	Comp. 5	10	150	90	No luminescence
6	Comp. 6	10	290	200	40
7	Comp. 7	10	190	160	20
8	Comp. 8	10	320	240	80

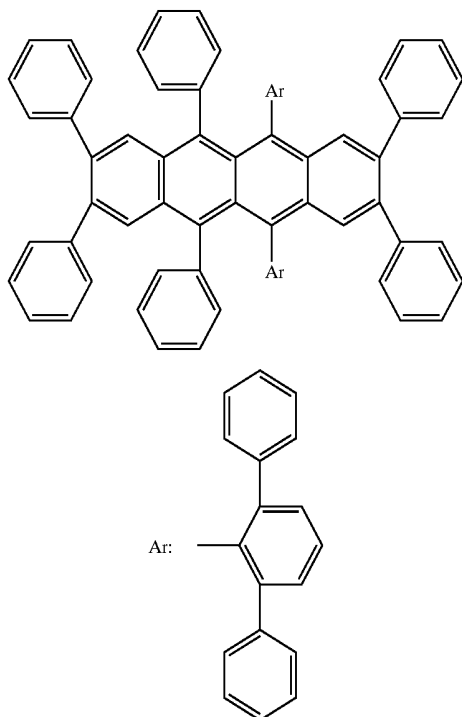
Comparative compound No. 7

25



Comparative compound No. 8

40



EXAMPLE 16

An organic luminescence device shown in FIG. 3 was prepared in the following manner.

In a similar-manner as in Example 1, on a transparent electroconductive support, a 120 nm-thick ITO film (anode) and a 30 nm-thick hole transport layer 5 were formed.

On the hole transport layer 5, a 20 nm-thick luminescence layer 3 of a fused polynuclear compound (Ex. Comp. No. 4) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

On the luminescence layer 3, a 40 nm-thick electron transport layer 6 of tris-(8-hydroxy-quinoline)aluminum (Alq3) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

Then, on the electron transport layer 6, a 150 nm-thick metal electrode (cathode 4) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa; 1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 8 volts was applied between the ITO electrode (anode 2, positive pole) and the Al—Li electrode (cathode 4, negative pole), whereby a current was passed through the

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organic luminescence device at a current density of 8.1 mA/cm² and blue luminescence was observed at a luminance of 3980 cd/m².

Then, when the organic luminescence device was supplied with a voltage for 100 hours while keeping a current density of 7.0 mA/cm² in a nitrogen atmosphere, a luminance of 3090 cd/m² (as initial luminance) was merely decreased to 2600 cd/m² even after 100 hours of the voltage application, thus exhibiting a good durability. The results are also shown in Table 3 appearing hereinafter.

EXAMPLES 17-30

Organic luminescence devices were prepared and evaluated in the same manner as in Example 16 except that the fused polynuclear compound (Ex. Comp. No. 4) was changed to those (Ex. Comp. Nos. 6, 11, 14, 18, 22, 27, 29, 35, 40, 42, 47, 49, 52 and 62), respectively.

The results are shown in Table 3.

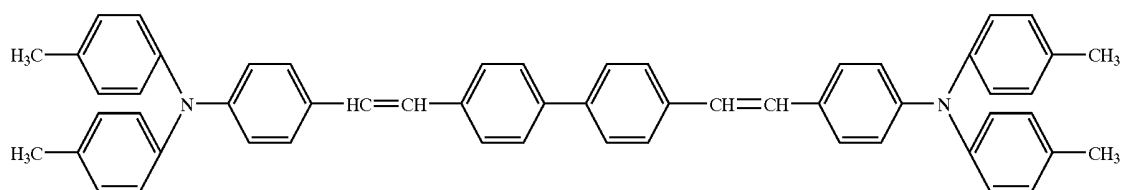
COMPARATIVE EXAMPLES 9-16

Organic luminescence devices were prepared and evaluated in the same manner as in Example 16 except that the fused polynuclear compound (Ex. Comp. No. 4) was changed to the above-mentioned comparative compounds Nos. 1-8, respectively.

The results are shown in Table 4.

TABLE 3

Ex. No.	Comp. No.	Applied voltage (V)	Initial Luminance (at 7.0 mA/cm ²)		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
16	4	8	3980	3090	2600
17	6	8	4140	3470	2480
18	11	8	7370	6510	4860
19	14	8	5320	4130	2990
20	18	8	7050	6600	5210
21	22	8	5560	4430	3310
22	27	8	5920	4790	3600
23	29	8	7760	7000	4940
24	35	8	3170	2770	2030
25	40	8	5160	4410	3200
26	42	8	6010	5300	4290
27	47	8	6990	6570	5100
28	49	8	7500	6380	5290
29	52	8	5300	4680	3800
30	62	8	4950	4000	3350



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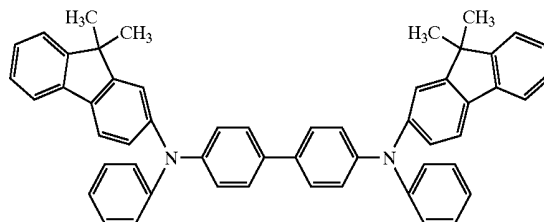
TABLE 4

Comp. Ex. No.	Comp. No.	Applied voltage (V)	Initial Luminance (at 7.0 mA/cm ²)		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
9	Comp. 1	8	640	450	60
10	Comp. 2	8	470	430	No luminescence
11	Comp. 3	8	340	270	No luminescence
12	Comp. 4	8	490	420	No luminescence
13	Comp. 5	8	650	490	No luminescence
14	Comp. 6	8	1700	1000	140
15	Comp. 7	9	1180	860	90
16	Comp. 8	8	2120	1240	360

EXAMPLE 31

An organic luminescence device shown in FIG. 3 was prepared in the following manner.

On a transparent electroconductive support prepared in the same manner as in Example 1, a solution of a hole transport material shown below in chloroform was applied by spin coating to form a 20 nm-thick hole transport layer 5.



On the hole transport layer 5, a 20 nm-thick co-deposited luminescence layer 3 of compound shown below fused polynuclear compound (Ex. Comp. No. 2) (1/50 by weight) was formed by vacuum deposition (1.0×10⁻⁴ Pa; 0.2-0.3 nm/sec).

On the luminescence layer **3**, a 40 nm-thick electron transport layer **6** of tris-18-hydroxy-quinoline)aluminum (Alq3) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

Then, on the electron transport layer **6**, a 150 nm-thick metal electrode (cathode **4**) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa; 1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 8 volts was applied between the ITO electrode (anode **2**, positive pole) and the Al—Li electrode (cathode **4**, negative pole), whereby a current was passed through the organic luminescence device at a current density of 8.5 mA/cm² and bluish white luminescence was observed at a luminance of 46500 cd/m².

Then, when the organic luminescence device was supplied with a voltage for 100 hours while keeping a current density of 5.0 mA/cm² in a nitrogen atmosphere; a luminance of 22500 cd/m² (as initial luminance) was merely decreased to 17600 cd/m² even after 100 hours of the voltage application, thus exhibiting a good durability. The results are also shown in Table 5 appearing hereinafter.

EXAMPLES 32–45

Organic luminescence devices were prepared and evaluated in the same manner as in Example 31 except that the fused polynuclear compound (Ex. Comp. No. 2) was changed to those (Ex. Comp. Nos. 7, 9, 15, 17, 23, 28, 32, 34, 38, 41, 45, 50, 53 and 56), respectively.

The results are shown in Table 5.

COMPARATIVE EXAMPLES 17–24

Organic luminescence devices were prepared and evaluated in the same manner as in Example 31 except that the fused polynuclear compound (Ex. Comp. No. 2) was changed to the above-mentioned comparative compounds Nos. 1–8, respectively.

The results are shown in Table 6.

TABLE 5

Ex. No.	Comp. No.	Applied voltage (V)	Initial Luminance (at 5.0 mA/cm ²)		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
31	2	8	46500	22500	17600
32	7	8	24300	13500	10900
33	9	8	78300	39000	32100
34	15	8	66000	35400	29800
35	17	8	69600	37000	33300
36	23	8	45600	23000	18500
37	28	8	55200	29000	25600
38	32	8	67700	37000	30800
39	34	8	43200	24000	16500
40	38	8	41000	20000	15500
41	41	8	46700	25300	20900
42	45	8	59800	34100	25000
43	50	8	39700	26400	22000
44	53	8	62300	34000	28700
45	56	8	44300	26800	20800

TABLE 6

Comp. No.	Ex. No.	Applied voltage (V)	Initial Luminance (at 5.0 mA/cm ²)		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
17	Comp. 1	8	1190	650	130
18	Comp. 2	8	770	460	No luminescence
19	Comp. 3	8	650	390	No luminescence
20	Comp. 4	8	690	390	No luminescence
21	Comp. 5	8	1250	880	140
22	Comp. 6	8	5700	2610	870
23	Comp. 7	8	3180	1820	760
24	Comp. 8	8	7220	3540	1360

EXAMPLE 46

An organic luminescence device shown in FIG. 5 was prepared in the following manner.

On a transparent electroconductive support prepared in the same manner as in Example 1, a 20 nm-thick hole transport layer **5** was formed in the same manner as in Example 31.

On the hole transport layer **5**, a 20 nm-thick co-deposited luminescence layer **3** of rubrene/Alq3 (1:20 by weight) was formed by vacuum deposition (1.0×10^{-4} Pa 0.2–0.3 nm/sec).

On the luminescence layer **3**, a 10 nm-thick hole/exciton blocking layer **8** of a fused polynuclear compound (Ex. Comp. No. 3) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

On the blocking layer **8**, a 40 nm-thick electron transport layer **6** of Alq3 was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

Then, on the electron transport layer **6**, a 150 nm-thick metal electrode (cathode **4**) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa; 1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 10 volts was applied between the ITO electrode (anode **2**, positive pole) and the Al—Li electrode (cathode **4**, negative pole), whereby a current was passed through the organic luminescence device at a current density of 8.9 mA/cm² and yellowish green luminescence was observed at a luminance of 60200 cd/m².

Then, when the organic luminescence device was supplied with a voltage for 100 hours while keeping a current density of 7.0 mA/cm² in a nitrogen atmosphere, a luminance of 38000 cd/m² (as initial luminance) was merely decreased to 28700 cd/m² even after 100 hours of the voltage application, thus exhibiting a good durability. The results are also shown in Table 7 appearing hereinafter.

EXAMPLES 47–60

Organic luminescence devices were prepared and evaluated in the same manner as in Example 46 except that the fused polynuclear compound (Ex. Comp. No. 3) was changed to those (Ex. Comp. Nos. 8, 13, 21, 30, 33, 37, 44, 48, 54, 55, 57, 59, 60 and 61), respectively.

The results are shown in Table 7.

COMPARATIVE EXAMPLES 25–32

Organic luminescence devices were prepared and evaluated in the same manner as in Example 46 except that the

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fused polynuclear compound (Ex. Comp. No. 3) was changed to the above-mentioned comparative compounds Nos. 1–8, respectively.

The results are shown in Table 8.

TABLE 7

Ex. No.	Comp. No.	Applied voltage (V)	Initial		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
46	3	10	60200	38000	28700
47	8	10	41000	24500	15500
48	13	10	62400	39300	30100
49	21	10	61100	38500	29900
50	30	10	79700	47000	34100
51	33	10	77600	45800	31200
52	37	10	42500	26000	15800
53	44	10	67700	37000	30800
54	48	10	80200	47000	34400
55	54	10	42900	24200	16500
56	55	10	76600	45300	31800
57	57	10	49900	27300	15000
58	59	10	39200	24400	15700
59	60	10	42500	28000	18700
60	61	10	42300	26300	17000

TABLE 8

Comp. Ex.	Comp. No.	Applied voltage (V)	Initial		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
25	Comp. 1	10	1020	660	120
26	Comp. 2	10	750	420	No luminescence
27	Comp. 3	10	680	410	No luminescence
28	Comp. 4	10	690	420	No luminescence
29	Comp. 5	10	1050	750	210
30	Comp. 6	10	5400	2200	770
31	Comp. 7	10	2850	1600	560
32	Comp. 8	10	8010	3670	910

EXAMPLE 61

An organic luminescence device shown in FIG. 1 was prepared in the following manner.

On a transparent electroconductive support prepared in the same manner as in Example 1, a solution of a mixture comprising 0.050 g of a fused polynuclear compound (Ex. Comp. No. 12) and 1.00 g of poly-N-vinylcarbazole (weight-average molecular weight=63,000) in 80 ml of chloroform was applied by spin coating (2000 rpm) to form a 120 nm-thick luminescence layer 3.

Then, on the luminescence layer 3, a 150 nm-thick metal electrode (cathode 4) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa; 1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 10 volts was applied between the ITO electrode (anode 2, positive pole) and the Al—Li electrode (cathode 4, negative pole), whereby a current was passed through the organic luminescence device at a current density of 7.8 mA/cm² and blue luminescence was observed at a luminance of 1250 cd/m².

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Then, when the organic luminescence device was supplied with a voltage for 100 hours while keeping a current density of 5.0 mA/cm² in a nitrogen atmosphere, a luminance of 820 cd/m² (as initial luminance) was merely decreased to 670 cd/m² even after 100 hours of the voltage application, thus exhibiting a good durability. The results are also shown in Table 9 appearing hereinafter.

EXAMPLES 62 and 63

Organic luminescence devices were prepared and evaluated in the same manner as in Example 61 except that the fused polynuclear compound (Ex. Comp. No. 12) was changed to those (Ex. Comp. Nos. 20 and 26), respectively. The results are shown in Table 9.

COMPARATIVE EXAMPLES 33–40

Organic luminescence devices were prepared and evaluated in the same manner as in Example 61 except that the fused polynuclear compound (Ex. Comp. No. 12) was changed to the above-mentioned comparative compounds Nos. 1–8, respectively.

The results are shown in Table 9.

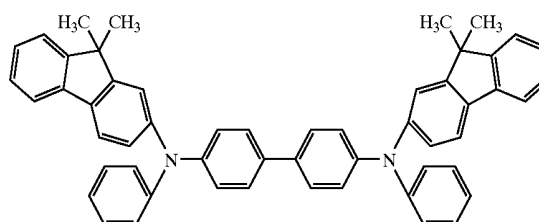
TABLE 9

Ex. or Comp. Ex.	Ex. or Comp. No.	Applied voltage (V)	Initial		
			Luminance (cd/m ²)	Initial (cd/m ²)	After 100 hr (cd/m ²)
30	Ex. 61	10	1250	820	670
	Ex. 62	10	980	670	590
	Ex. 63	10	1070	740	610
	Comp. Ex. 33	10	230	150	No luminescence
	Comp. Ex. 34	10	120	80	No luminescence
	Comp. Ex. 35	10	90	70	No luminescence
	Comp. Ex. 36	10	80	60	No luminescence
	Comp. Ex. 37	10	250	150	No luminescence
	Comp. Ex. 38	10	340	220	40
	Comp. Ex. 39	10	340	210	20
	Comp. Ex. 40	10	410	270	80

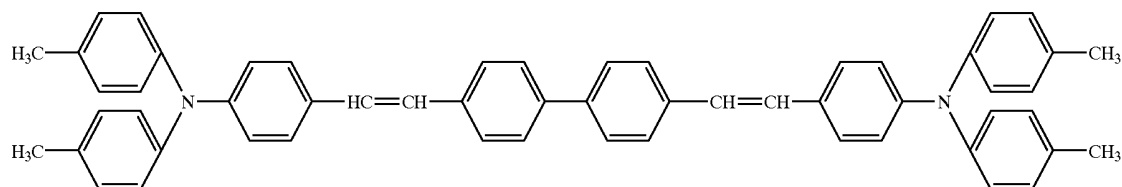
EXAMPLE 64

An organic luminescence device shown in FIG. 3 was prepared in the following manner.

On a transparent electroconductive support prepared in the same manner as in Example 1, a mixture of hole transport material shown below with rubrene (yellow luminescence material) (100/1 by weight) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec) to form a 50 nm-thick hole transport layer 5.



On the hole transport layer **5**, a 20 nm-thick luminescence layer **3** of a mixture of a compound shown below with a fused polynuclear compound (Ex. Comp. No. 9) ($1/_{100}$ by weight) was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).



On the luminescence layer **3**, a 30 nm-thick electron transport layer **6** of Alq3 was formed by vacuum deposition (1.0×10^{-4} Pa; 0.2–0.3 nm/sec).

Then, on the electron transport layer **6**, a 150 nm-thick metal electrode (cathode **4**) of an aluminum-lithium alloy (Li content: 1 atomic %) was formed by vacuum deposition (1.0×10^{-4} Pa; 1.0–1.2 nm/sec).

To the thus-prepared organic luminescence device, a DC voltage of 8 volts was applied between the ITO electrode (anode **2**, positive pole) and the Al—Li electrode (cathode **4**, negative pole), whereby a current was passed through the organic luminescence device at a current density of 100 mA/cm² and white luminescence was observed at a luminance of 11000 cd/m².

Then, when the organic luminescence device was supplied with a DC voltage of 15 volts, a current was passed through the organic luminescence device at a current density of 2250 mA/cm² and white luminescence was observed at a luminance of 213000 cd/m².

EXAMPLES 65–67

Organic luminescence devices were prepared and evaluated in the same manner as in Example 1 except that the fused polynuclear compound (Ex. Comp. No. 1) was changed to those (Ex. Comp. Nos. 64, 65 and 66), respectively.

The results are shown in Table 10.

TABLE 10

Ex. No.	Comp. No.	Applied voltage (V)	Luminance (at 7.0 mA/cm ²)		
			Initial (cd/m ²)	After 100 hr (cd/m ²)	
65	64	10	1900	1500	1400
66	65	10	2400	1800	1600
67	66	10	1500	1400	1200

EXAMPLES 68–70

Organic luminescence devices were prepared and evaluated in the same manner as in Example 16 except that the fused polynuclear compound (Ex. Comp. No. 4) was changed to those (Ex. Comp. Nos. 64, 65 and 66), respectively.

The results are shown in Table 11.

TABLE 11

Ex. No.	Comp. No.	Applied voltage (V)	Luminance (at 7.0 mA/cm ²)		
			Initial (cd/m ²)	After 100 hr (cd/m ²)	
68	64	8	11000	9000	7000
69	65	8	14000	12500	9000
70	66	8	9500	8500	7000

EXAMPLES 71–74

The organic luminescence devices prepared in Examples 21, 68, 69 and 70 were subjected to measurement off CIE chromaticity coordinates (X, Y) by using an emission spectrometer ("Photal MCPD-7000", mfd. by Otsuka Electronics. Co. Ltd.).

The results are shown in Table 12.

TABLE 12

Ex. No.	Ex. Comp. No.	CIE chromaticity coordinates (X, Y)
71	22	(0.14, 0.08)
72	64	(0.16, 0.12)
73	65	(0.15, 0.09)
74	66	(0.16, 0.13)

From the above results, it has been found that the fused polynuclear compounds (Ex. Comp. Nos. 22, 64, 65 and 66) used in the present invention were effective to cause blue luminescence with excellent purity.

As described hereinabove, according to the present invention, by using a fused polynuclear compound of the formula (I) to (VII) as a material for organic luminescence function layer, particularly for an electron transport layer, a luminescence layer or a hole/exciton blocking layer, in an organic luminescence device, the resultant organic luminescence device allows a high-luminance luminescence at a lower applied voltage and is also excellent in durability.

The organic luminescence device of the present invention can be readily prepared by vacuum deposition or wet coating, thus being produced in a large-area device relatively inexpensively.

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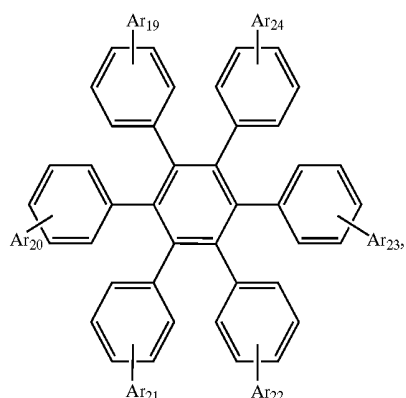
What is claimed is:

1. A device, comprising:

a pair of an anode and a cathode, and

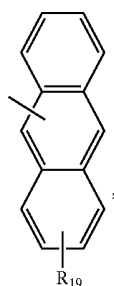
at least one organic luminescence layer disposed between the anode and the cathode,

wherein said at least one organic luminescence layer comprises a layer of a compound represented by the following formula (VII):



wherein each one of Ar₁₉, Ar₂₀, Ar₂₁, Ar₂₂, Ar₂₃ and Ar₂₄ is the same substituted or unsubstituted fused polynuclear carbocyclic aromatic group or substituted or unsubstituted acrydinyl or fluorenyl group.

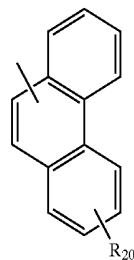
2. The device according to claim 1, wherein Ar₁₉, Ar₂₀, Ar₂₁, Ar₂₂, Ar₂₃ and Ar₂₄ are selected from the group consisting of formulas (VIII), (IX), (X) and (XIII):



wherein R₁₉ is a hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or a cyano group;

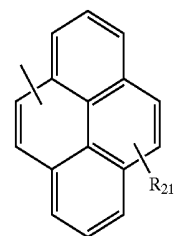
76

(IX)



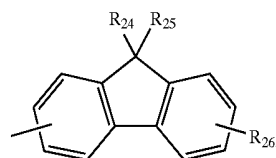
wherein R₂₀ is a hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or a cyano group;

(X)



wherein R₂₁ is a hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or a cyano group; or

(XIII)



wherein R₂₄, R₂₅ and R₂₆ independently denote a hydrogen atom, an alkyl group, a substituted or unsubstituted aralkyl group, a substituted or unsubstituted aryl group, a substituted or unsubstituted heterocyclic group, a substituted amino group or a cyano group.

* * * * *

UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 6,994,922 B2
APPLICATION NO. : 10/940734
DATED : February 7, 2006
INVENTOR(S) : Koichi Suzuki et al.

Page 1 of 3

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

ON COVER PAGE AT (57) ABSTRACT

Line 7, “denote hydrogen” should read --denote a hydrogen--.

COLUMN 1

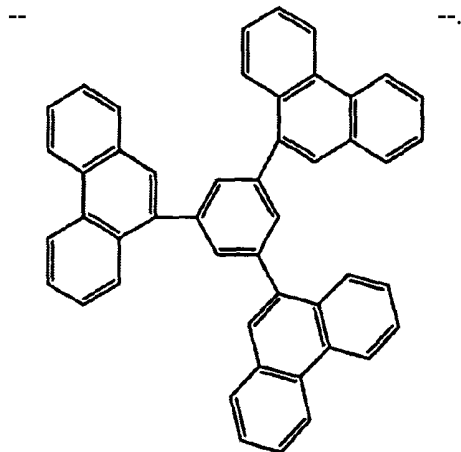
Line 36, “5,151,629” should read --5,151,629;--.

COLUMN 3

Line 58, “br” should read --or--.

COLUMN 14

Figure 14, “” should read



UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 6,994,922 B2
APPLICATION NO. : 10/940734
DATED : February 7, 2006
INVENTOR(S) : Koichi Suzuki et al.

Page 2 of 3

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

COLUMN 41

Line 60, "formed-in" should read --formed in--.

COLUMN 43

Line 5, "confining" should read --confine--; and

Line 7, "luminescence-efficiency." should read --luminescence efficiency.--.

COLUMN 61

Line 65, "leaning" should read --cleaning--.

COLUMN 66

Line 47, "similar-manner" should read --similar manner--.

COLUMN 69

Line 2, "tris-18-hydroxy-quinoline)aluminum" should read --tris-(8-hydroxy-quinoline)aluminum--.

COLUMN 71

Line 2, "the:above-mentioned" should read --the above-mentioned--.

COLUMN 72

Line 13, "to-those" should read --to those--.

UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 6,994,922 B2
APPLICATION NO. : 10/940734
DATED : February 7, 2006
INVENTOR(S) : Koichi Suzuki et al.

Page 3 of 3


It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

COLUMN 75

Line 29, "carbocylic" should read --carbocyclic--.

Signed and Sealed this

Fourteenth Day of November, 2006

A handwritten signature in black ink on a light gray dotted background. The signature is written in a cursive style and reads "Jon W. Dudas".

JON W. DUDAS

Director of the United States Patent and Trademark Office

专利名称(译)	具有稠合多核化合物的有机发光装置		
公开(公告)号	US6994922	公开(公告)日	2006-02-07
申请号	US10/940734	申请日	2004-09-15
[标]申请(专利权)人(译)	佳能株式会社		
申请(专利权)人(译)	佳能株式会社		
当前申请(专利权)人(译)	佳能株式会社		
[标]发明人	SUZUKI KOICHI SENOO AKIHIRO TANABE HIROSHI		
发明人	SUZUKI, KOICHI SENOO, AKIHIRO TANABE, HIROSHI		
IPC分类号	H05B33/14 H01L51/50 C07C13/547 C07C13/567 C09K11/06 H01L51/00 H01L51/30		
CPC分类号	C07C13/567 C09K11/06 H01L51/005 H01L51/0054 H01L51/0055 H01L51/0056 H01L51/0058 H01L51/0059 H01L51/006 H01L51/0067 H01L51/0068 H01L51/0069 H01L51/0072 H01L51/0095 Y10S428/917 C09K2211/1003 C09K2211/1007 C09K2211/1011 C09K2211/1014 C09K2211/1029 C09K2211/1037 C09K2211/1092 H01L51/0042 H01L51/0081 H01L51/5012 H01L2251/308 C07C2103/18 C07C2603/18		
优先权	2001046225 2001-02-22 JP 2002036804 2002-02-14 JP		
其他公开文献	US20050048318A1		
外部链接	Espacenet USPTO		

摘要(译)

一种有机发光器件，具有阳极和阴极以及至少一个设置在阳极和阴极之间的有机发光功能层。上述至少一个有机发光功能层包括由下式 (I) 表示的稠合多核化合物层：其中R1，R2，R3和R4独立地表示氢原子，烷基，取代或未取代的芳烷基，取代或未取代的芳基，取代或未取代的杂环基，取代的氨基或氰基；Ar1和Ar2独立地表示取代或未取代的稠合多核芳香族基团或取代或未取代的稠合多核杂环基。

