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(54) **MATERIALS FOR ORGANIC ELECTROLUMINESCENT DEVICES**

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CPC ***C07C 43/275*** (2013.01); ***C07C 13/567*** (2013.01); ***C07C 15/28*** (2013.01); ***C07C 15/52*** (2013.01); ***C07C 211/54*** (2013.01); ***C07C 211/56*** (2013.01); ***C07C 211/58*** (2013.01); ***C09K 11/06*** (2013.01); ***H01L 51/0052*** (2013.01); ***H01L 51/0058*** (2013.01); ***H01L 51/006*** (2013.01); ***H05B 33/14*** (2013.01); *C09K 2211/1011* (2013.01); *H01L 51/0071* (2013.01); *H01L 51/0072* (2013.01); *H01L 51/0094* (2013.01); *H01L 51/5012* (2013.01); *H01L 51/5048* (2013.01); *Y02E 10/549* (2013.01); *Y10S 428/917* (2013.01)

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See application file for complete search history.

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(57) **ABSTRACT**

The present invention relates to anthracene derivatives, to the use thereof in organic electroluminescent devices, and to organic electroluminescent devices comprising these compounds.

14 Claims, No Drawings

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MATERIALS FOR ORGANIC ELECTROLUMINESCENT DEVICES

RELATED APPLICATIONS

This application is a national stage application (under 35 U.S.C. §371) of PCT/EP2007/001732, filed Feb. 28, 2007, which claims benefit of German application 102006013802.3, filed Mar. 24, 2006.

The present invention relates to novel anthracene derivatives, to the use thereof in organic electroluminescent devices, and to organic electroluminescent devices comprising these compounds.

Organic semiconductors are used as functional materials in a number of different applications which can be ascribed to the electronics industry in the broadest sense. The general structure of organic electroluminescent devices which are capable of the emission of light in the visible spectral region is described, for example, in U.S. Pat. No. 4,539,507, U.S. Pat. No. 5,151,629, EP 0676461 and WO 98/27136.

However, these devices still exhibit considerable problems which require urgent improvement for use in high-quality full-colour displays:

1. The efficiency, colour and lifetime of the organic electroluminescent devices are still inadequate for high-quality applications.
2. The compounds used frequently do not have a sufficiently high glass-transition temperature.
3. The redox stability (stability to holes and electrons) of the compounds used to date is still inadequate.
4. The charge-carrier mobility, in particular the electron mobility, is inadequate.
5. The operating voltage should be reduced still further, in particular for mobile applications.

The closest prior art which may be mentioned is the use of various condensed aromatic compounds, in particular anthracene or pyrene derivatives, as host materials, in particular for blue-emitting electroluminescent devices. 9,10-Bis(2-naphthyl)anthracene is known as host material in accordance with the prior art (U.S. Pat. No. 5,935,721). Further anthracene derivatives which are suitable as host materials are described in WO 01/076323, in WO 01/021729, in WO 04/013073, in WO 04/018588, in WO 03/087023 or in WO 04/018587. Host materials based on aryl-substituted pyrenes and chrysenes are described in WO 04/016575. WO 03/095445 and CN 1362464 describe 9,10-bis(1-naphthyl) anthracene derivatives for use in OLEDs.

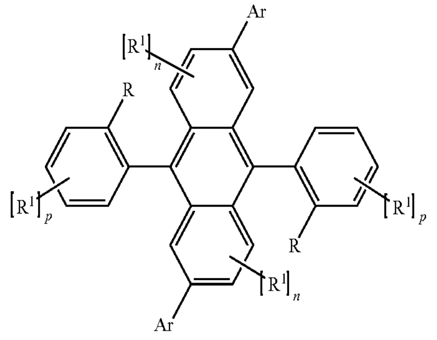
The object of the present invention was to provide compounds having improved properties, in particular improved host materials.

Surprisingly, it has been found that organic electroluminescent devices which comprise anthracene derivatives which are substituted in the 9,10-position by ortho-substituted phenyl groups and which are furthermore substituted in the 2,6-position by aryl or heteroaryl groups have significant improvements compared with the prior art. The present invention therefore relates to these compounds and to the use thereof in OLEDs.

WO 03/060956 and WO 05/097756 disclose anthracene derivatives which are substituted in the 9,10-position by ortho-biphenyl and which are furthermore substituted in the 2,6-position by aryl groups which contain benzimidazole. The positive effect of these compounds is attributed to the presence of the benzimidazole groups.

The invention relates to compounds of the formula (1)

Formula (1)



where the following applies to the symbols and indices used:

Ar is on each occurrence, identically or differently, an aromatic or heteroaromatic ring system having 6 to 30 aromatic ring atoms, which may be substituted by one or more radicals R^1;

R, R^1 are, identically or differently on each occurrence, F, Cl, Br, I, CN, $N(Ar^1)_2$, $C(=O)Ar^1$, $P(Ar^1)_2$, $P(—O)(Ar^1)_2$, $Si(R^2)_3$, NO_2, a straight-chain alkyl, alkoxy or thioalkoxy group having 1 to 40 C atoms or a branched or cyclic alkyl, alkoxy or thioalkoxy group having 3 to 40 C atoms, each of which may be substituted by one or more radicals R^2, where one or more non-adjacent CH_2 groups may be replaced by $—R^2C=CR^2$, $—C\equiv C—$, $Si(R^2)_2$, $Ge(R^2)_2$, $Sn(R^2)_2$, $C=$, $C=S$, $C=Se$, $C=NR^2$, —O—, —S—, $—N(R^2)—$ or $—CONR^2—$ and where one or more H atoms may be replaced by F, Cl, Br, I, CN or NO_2, or an aromatic or heteroaromatic ring system having 5 to 30 aromatic ring atoms, which may be substituted by one or more radicals R^2 or an aryloxy or heteroaryloxy group having 5 to 24 aromatic ring atoms, which may be substituted by one or more radicals R^2, or a combination of two, three, four or five of these systems; adjacent substituents R and R^1 or adjacent substituents R^1 here may also form a mono- or polycyclic, aliphatic ring system with one another;

Ar^1 is on each occurrence, identically or differently, an aromatic or heteroaromatic ring system having 5 to 30 aromatic ring atoms, which may be substituted by one or more non-aromatic radicals R^1; two radicals Ar^1 here may also be connected to one another by a single bond or an O, S, $N(R^2)$ or $C(R^2)_2$ group;

R^2 is on each occurrence, identically or differently, H or a hydrocarbon radical having 1 to 20 C atoms, which may be aliphatic or aromatic or a combination of aliphatic and aromatic and which may also be substituted by F; two or more radicals R^2 here may also form a mono- or polycyclic, aliphatic or aromatic ring system with one another;

n is, identically or differently on each occurrence, 0, 1, 2 or 3;

p is, identically or differently on each occurrence, 0, 1, 2, 3 or 4;

with the proviso that the substituents Ar are not substituted or unsubstituted benzimidazole and that no radicals R^1 which contain substituted or unsubstituted benzimidazole are bonded to the substituents Ar.

For the purposes of this invention, an aryl group or heteroaryl group is taken to mean an aromatic group or heteroaromatic group respectively having a common aromatic π-electron system. For the purposes of this invention, this may be a simple homo- or heterocycle, for example benzene, pyridine, etc., or it may be a condensed aryl or heteroaryl group in which at least two aromatic or heteroaromatic rings, for example benzene rings, are "fused" to one another, i.e. are condensed onto one another by anellation, i.e. have at least one common edge and thus also a common aromatic π-electron system. These aryl or heteroaryl groups may be substituted or unsubstituted; any substituents present may likewise form further ring systems. Thus, for example, systems such as naphthalene, anthracene, phenanthrene, pyrene, etc., are to be regarded as acyl groups and quinoline, acridine, benzothiophene, carbazole, etc., are as heteroaryl groups for the purposes of this invention, while, for example, biphenyl, fluorene, spirobifluorene, etc., are not aryl groups since they involve separate aromatic electron systems.

For the purposes of this invention, an aromatic ring system contains 6 to 40 C atoms in the ring system. For the purposes of this invention, a heteroaromatic ring system contains 2 to 40 C atoms and at least one heteroatom in the ring system, with the proviso that the total number of C atoms and heteroatoms is at least 5. The heteroatoms are preferably selected from N, O and/or S. These aromatic and heteroaromatic ring systems may be substituted or unsubstituted; any substituents present may likewise form further ring systems. For the purposes of this invention, an aromatic or heteroaromatic ring system is intended to be taken to mean a system which does not necessarily contain only aryl or heteroaryl groups, but instead in which a plurality of aryl or heteroaryl groups may also be interrupted by a short non-aromatic unit (preferably less than 10% of the atoms other than H), such as, for example, an sp^3-hybridised C, N or O atom. Thus, for example, systems such as 9,9'-spirobifluorene, fluorene, 9,9-diarylfluorene, triarylamine, diaryl ethers, etc., are also to be regarded as aromatic ring systems for the purposes of this invention. Part of the aromatic or heteroaromatic ring system here may also be a condensed group.

For the purposes of this invention, a cyclic alkyl group is taken to mean both monocyclic and bi- and polycyclic alkyl groups.

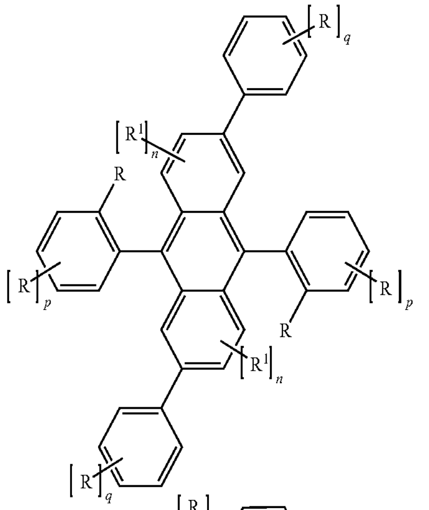
For the purposes of the present invention, a C_1- to C_{40}-alkyl group, in which, in addition, individual H atoms or CH_2 groups may be substituted by the above-mentioned groups, is particularly preferably taken to mean the radicals methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, s-butyl, t-butyl, 2-methylbutyl, n-pentyl, s-pentyl, cyclopentyl, n-hexyl, cyclohexyl, n-heptyl, cycloheptyl, n-octyl, cyclooctyl, 2-ethylhexyl, trifluoromethyl, pentafluoroethyl, 2,2,2-trifluoroethyl, ethenyl, propenyl, butenyl, pentenyl, cyclopentenyl, hexenyl, cyclohexenyl, heptenyl, cycloheptenyl, octenyl, cyclooctenyl, ethynyl, propynyl, butynyl, pentynyl, hexynyl or octynyl. A C_1- to C_{40}-alkoxy group is particularly preferably taken to mean methoxy, trifluoro-methoxy, ethoxy, n-propoxy, i-propoxy, n-butoxy, i-butoxy, s-butoxy, t-butoxy or 2-methylbutoxy. An aromatic or heteroaromatic ring system having 1 to 30 aromatic ring atoms, which may in each case also be substituted by the above-mentioned radicals R^1 and R^2 and which may be linked to the aromatic or heteroaromatic via any desired positions, is taken to mean, in particular, groups derived from benzene, naphthalene, anthracene, phenanthrene, pyrene, chrysene, perylene, fluoranthene, tetracene, pentacene, benzopyrene, biphenyl, biphenylene, terphenyl, terphenylene, fluorene, spirobifluorene, truxene, isotruxene, dihydrophenanthrene, dihydropyrene, tetrahydropyrene, cis- or trans-indenofluorene, furan, benzofuran, isobenzofuran, dibenzofuran, thiophene, benzothiophene, isobenzothiophene, dibenzothiophene, pyrrole, indole, isoindole, carbazole, pyridine, quinoline, isoquinoline, acridine, phenanthridine, benzo-5,6-quinoline, benzo-6,7-quinoline, benzo-7,8-quinoline, phenothiazine, phenoxazine, pyrazole, indazole, imidazole, benzimidazole, naphthimidazole, phenanthrimidazole, pyridimidazole, pyrazinimidazole, quinoxalinimidazole, oxazole, benzoxazole, naphthoxazole, anthroxazole, phenanthroxazole, isoxazole, 1,2-thiazole, 1,3-thiazole, benzothiazole, pyridazine, benzopyridazine, pyrimidine, benzopyrimidine, quinoxaline, pyrazine, phenazine, naphthyridine, azacarbazole, benzocarboline, phenanthroline, 1,2,3-triazole, 1,2,4-triazole, benzotriazole, 1,2,3-oxadiazole, 1,2,4-oxadiazole, 1,2,5-oxadiazole, 1,3,4-oxadiazole, 1,2,3-thiadiazole, 1,2,4-thiadiazole, 1,2,5-thiadiazole, 1,3,4-thiadiazole, 1,3,5-triazine, 1,2,4-triazine, 1,2,3-triazine, tetrazole, 1,2,4,5-tetrazine, 1,2,3,4-tetrazine, 1,2,3,5-tetrazine, purine, pteridine, indolizine, benzothiadiazole, triphenylamine, diphenylnaphthylamine, dinaphthylphenylamine, diphenyl ether, stilbene and tolan.

Preferred embodiments of compounds of the formula (1) are described below.

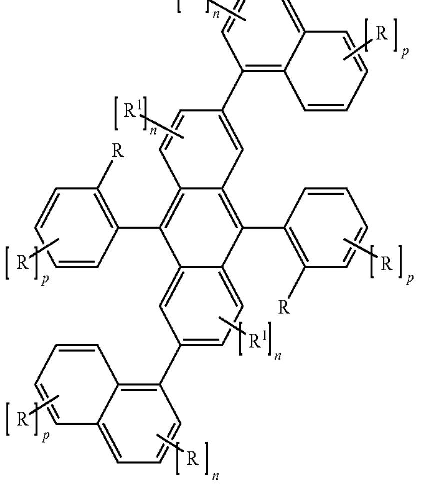
Preference is given to compounds of the formula (1) in which the symbol Ar stands for an aryl or heteroaryl group having 6 to 16 aromatic ring atoms, which may be substituted by R^1. The symbol Ar particularly preferably stands for phenyl, 2-pyridyl, 3-pyridyl, 4-pyridyl, 1-naphthyl, 2-naphthyl, 2-anthryl, 9-anthryl, 2-phenanthrenyl, 3-phenanthrenyl, 9-phenanthrenyl, 1-pyrenyl or 2-pyrenyl. The symbol Ar very particularly preferably stands for phenyl, 1-naphthyl, 2-naphthyl or 9-anthryl.

Both compounds of the formula (1) in which the two substituents Ar are selected identically and also compounds of the formula (1) in which the substituents Ar are different are in accordance with the invention. In a preferred embodiment of the invention, the two symbols Ar are selected identically. Particular preference is therefore given to the compounds of the formulae (2), (3), (4) and (5)

Formula (2)



Formula (3)



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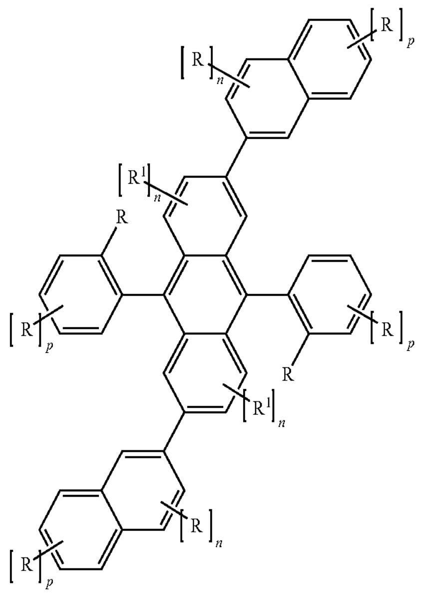
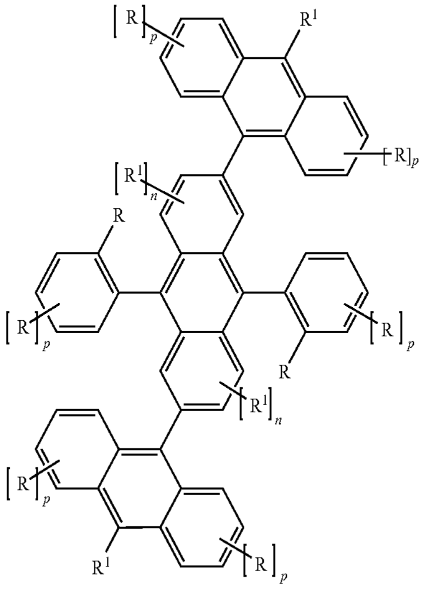
Formula (4)



Formula (5)



in which R, R^1, n and p have the same meaning as described above, and q stands for 0, 1, 2, 3, 4 or 5

In the compounds of the formulae (1) and (2) to (5), the phenyl groups in the 9- and 10-position on the anthracene may have hindered rotation about the anthracene-phenyl bond. For the purposes of this invention, hindered rotation is taken to mean a rotation barrier of at least 80 kJ/mol preferably at least 100 kJ/mol, in particular at least 120 kJ/mol at room temperature. This rotation barrier can be determined experimentally by temperature-dependent NMR measurements. If the compound of the formulae (1) and (2) to (5) exhibits atropisomerism about one or more bonds, the corresponding isolated or enriched atropisomers are in each case also a subject-matter of the invention. This relates both to enantiomers and also to diastereomers. Hindered rotation about the anthracene-phenyl bond is achieved by sufficiently large substituents R.

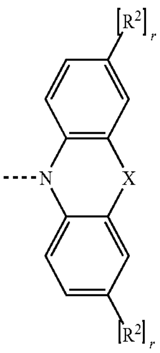
Preference is furthermore given to compounds of the formulae (1) and (2) to (5) in which the symbol R stands for $Si(R^2)_3$, $N(Ar^1)_2$, $C(=O)Ar^1$, $P(=O)(Ar^1)_2$, a straight-chain alkyl or alkoxy group having 1 to 10 C atoms or a branched or cyclic alkyl or alkoxy group having 3 to 10 C atoms, each of which may be substituted by one or more radicals R^2, where one or more non-adjacent CH_2 groups may be replaced by $—R^2C{=}CR^2—$ or —O— and where one or more H atoms may be replaced by F, or for an aryl or heteroaryl group having 5 to 16 aromatic ring atoms, which may be substituted by one or more radicals R^2, or a combination of two, three or four of these systems; adjacent substituents R and R^1 here may also form a mono- or polycyclic, aliphatic ring system with one another. R particularly preferably stands for $Si(R^2)_3$, a straight-chain alkyl group having 1 to 4 C atoms or a branched alkyl group having 3 to 5 C atoms, each of which may be substituted by one or more radicals R^2, where one or more H atoms may be replaced by F, or for an aryl or heteroaryl group having 6 to 14 aromatic ring atoms, which may be substituted by one or more radicals R^2, or a combination of two or three of these systems; adjacent substituents R and R^1 here may also form a mono- or polycyclic, aliphatic ring system with one another.

Both compounds of the formulae (1) and (2) to (5) in which the two substituents R are selected identically and also compounds of the formulae (1) and (2) to (5) in which the two substituents R are different are in accordance with the invention. The two substituents R are preferably selected identically.

Preference is furthermore given to compounds of the formulae (1) and (2) to (5) in which the symbol R^1, identically or differently on each occurrence, stands for $Si(R^2)_3$, F, $N(Ar^1)_2$, a straight-chain alkyl or alkoxy group having 1 to 6 C atoms or a branched or cyclic alkyl or alkoxy group having 3 to 10 C atoms, where in each case one or more CH_2 groups may be replaced by $—R^2C{=}CR^2—$ or —O— and where in each case one or more H atoms may be replaced by F, or an aryl or heteroaryl group having 5 to 14 aromatic ring atoms, which may be substituted by one or more radicals R^2, or a combination of two or three of these systems; two or more radicals R^1 here may also form a mono- or polycyclic, aliphatic ring system with one another or with an adjacent radical R. Particularly preferred radicals R^1 are selected from the group consisting of $Si(R^2)_3$, F, straight-chain alkyl groups having 1 to 4 C atoms or branched alkyl groups having 3 to 5 C atoms, where in each case one or more H atoms may be replaced by F, or aryl or heteroaryl groups having 6 to 10 aromatic ring atoms, or a combination of two of these systems; two or more adjacent radicals R^1 here may also form a mono- or polycyclic, aliphatic ring system with one another or with an adjacent radical R.

If one of the radicals R and/or R^1 stands for a group of the formula $N(Ar^1)_2$, it preferably stands for a group of the formula (6) or (7)

Formula (6)



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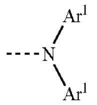
Formula (7)



where R^2 has the above-mentioned meaning and furthermore:

X stands for a single bond, O, S, $N(R^2)$ or $C(R^2)_2$;

Ar^1 is, identically or differently on each occurrence, an aryl or heteroaryl group having 5 to 20 aromatic ring atoms or a triarylamine group having 15 to 30 aromatic ring atoms, each of which may be substituted by one or more radicals R^1, preferably an aryl or heteroaryl group having 6 to 14 aromatic ring atoms or a triarylamine group having 18 to 26 aromatic ring atoms, each of which may be substituted by one or more non-aromatic radicals R^1, particularly preferably phenyl, o-tolyl, p-tolyl, o-fluorophenyl, p-fluorophenyl, 1-naphthyl, 2-naphthyl, triphenylamine or naphthyldiphenylamine;

r is on each occurrence, identically or differently, 0 or 1, preferably 0.

Preference is furthermore given to compounds of the formulae (1) and (2) to (5) in which the index n stands for 0 or 1, particularly preferably for 0.

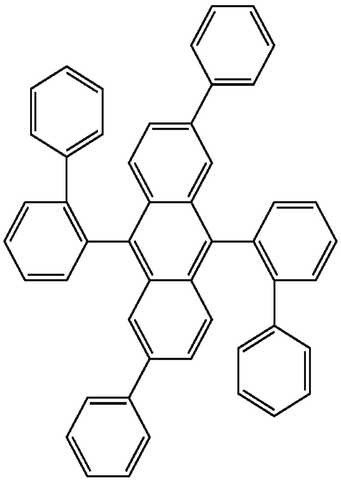
Preference is furthermore given to compounds of the formulae (1) and (2) to (5) in which the index p stands for 0, 1 or 2, particularly preferably for 0 or 1.

Preference is furthermore given to compounds of the formula (2) in which the index q stands for 0, 1, 2 or 3, particularly preferably for 0, 1 or 2, very particularly preferably for 0 or 1.

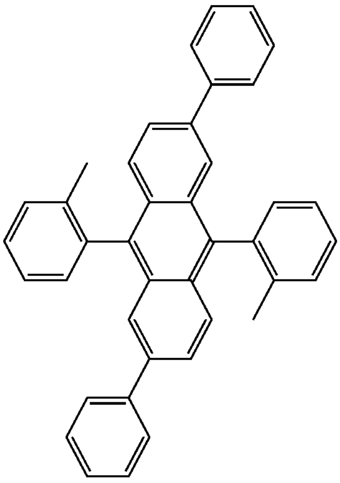
Preference is furthermore given to compounds of the formula (1) whose molecular weight is between 500 and 2000 g/mol, particularly preferably between 600 and 1500 g/mol.

Examples of preferred compounds of the formula (1) are compounds (1) to (50) depicted below.

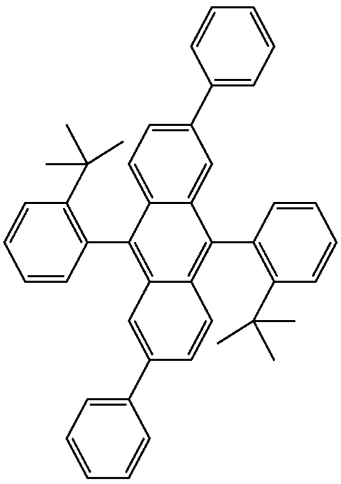
(1)



(2)



(3)



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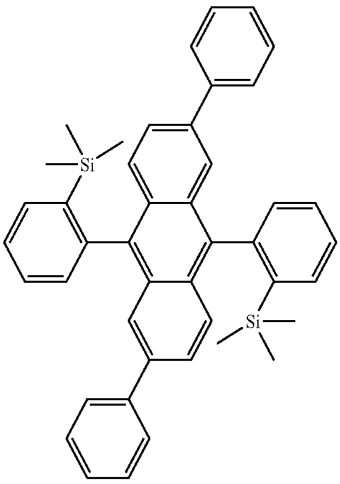

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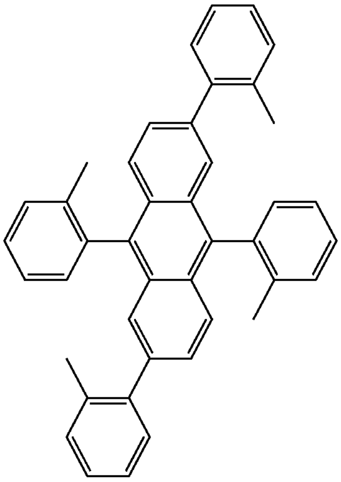

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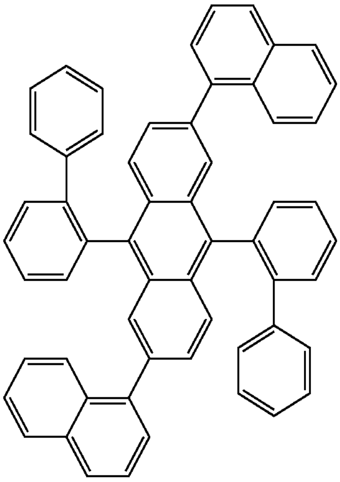

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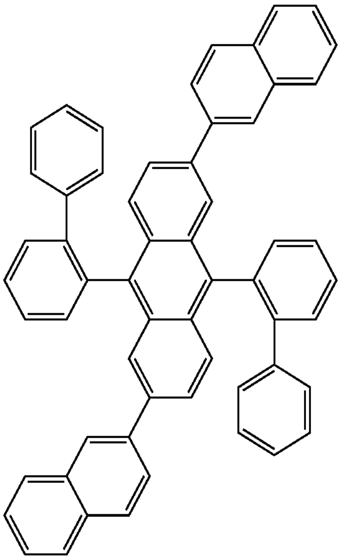

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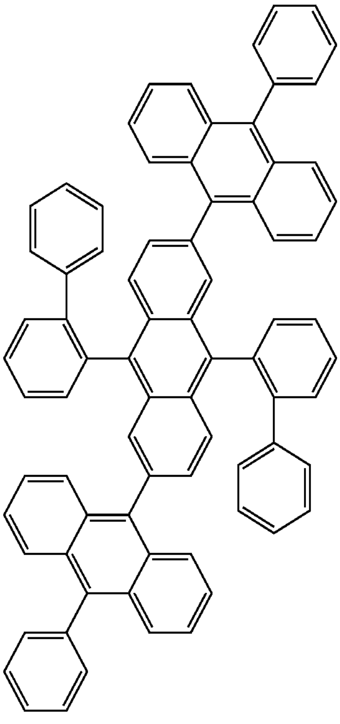

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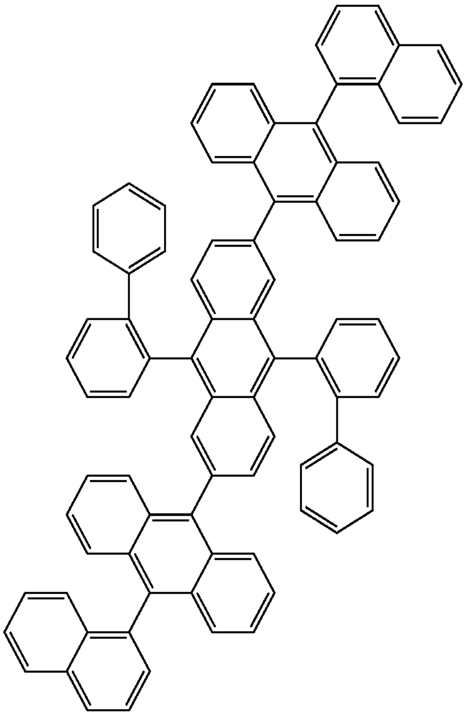

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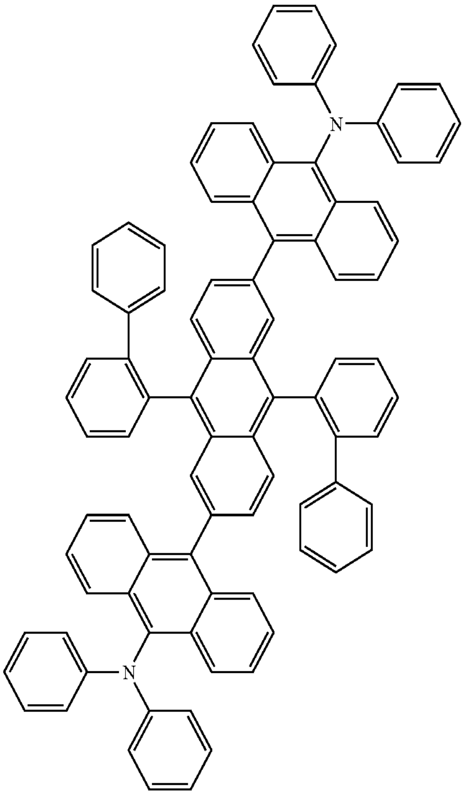

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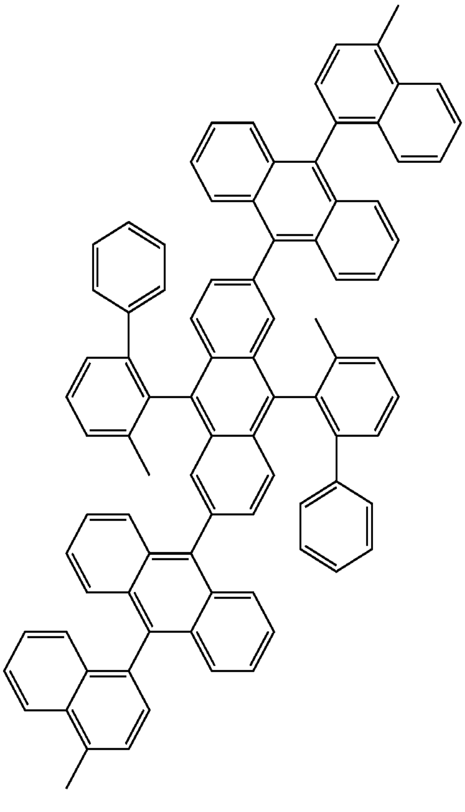

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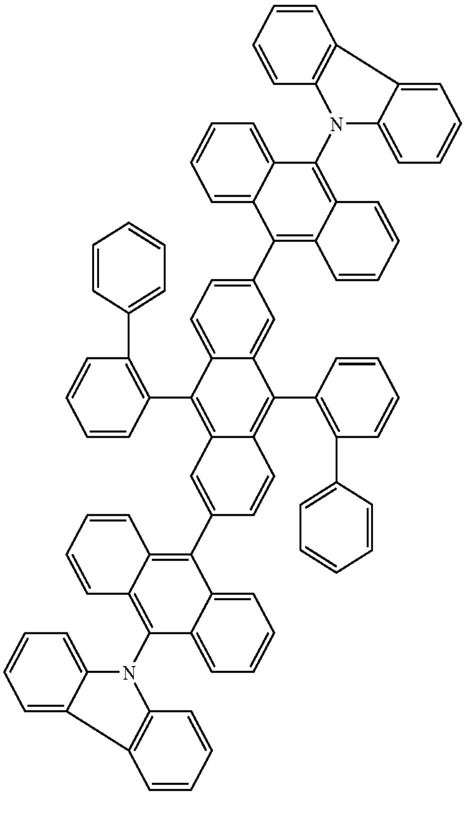

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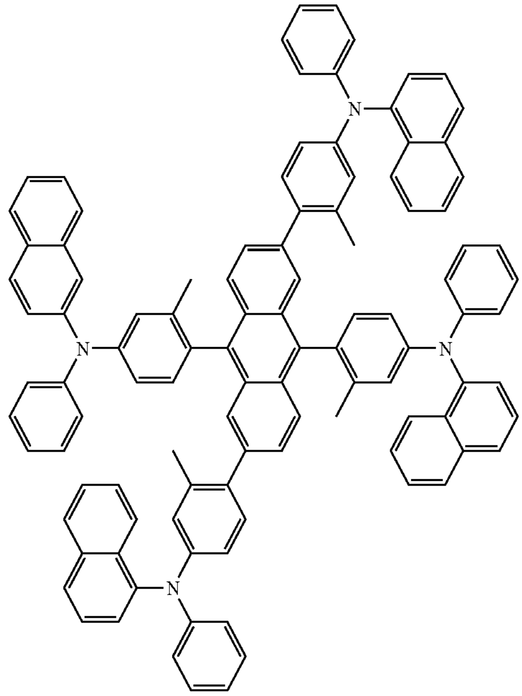

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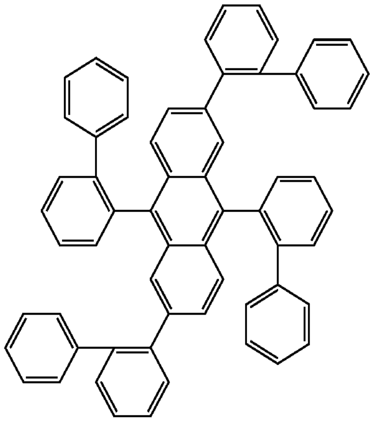

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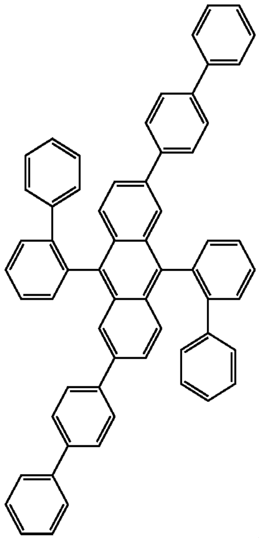

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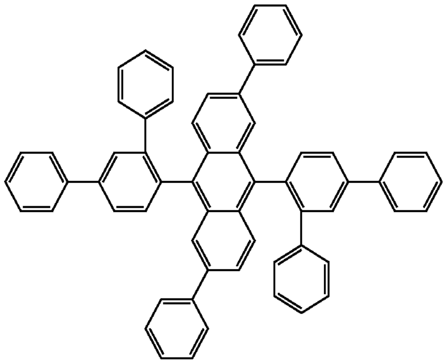

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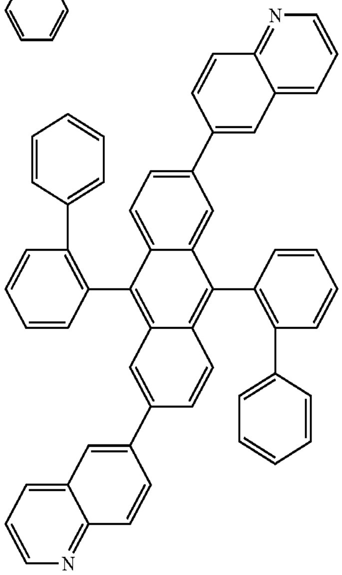

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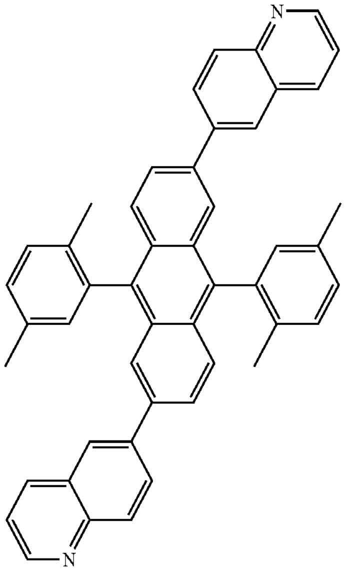

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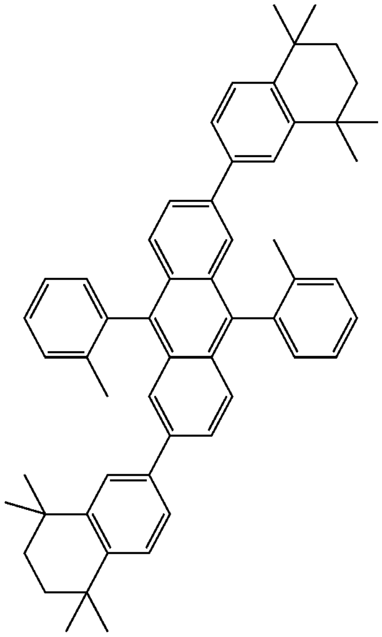

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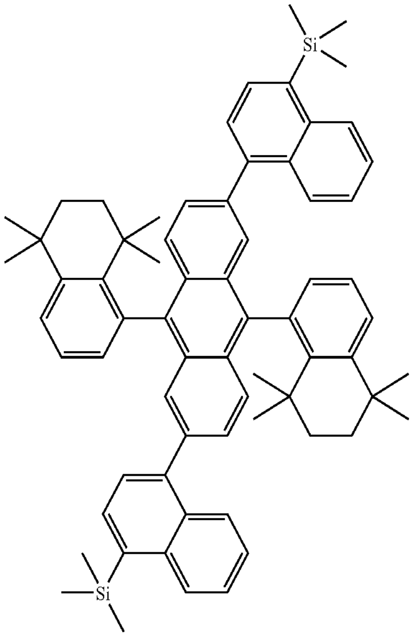

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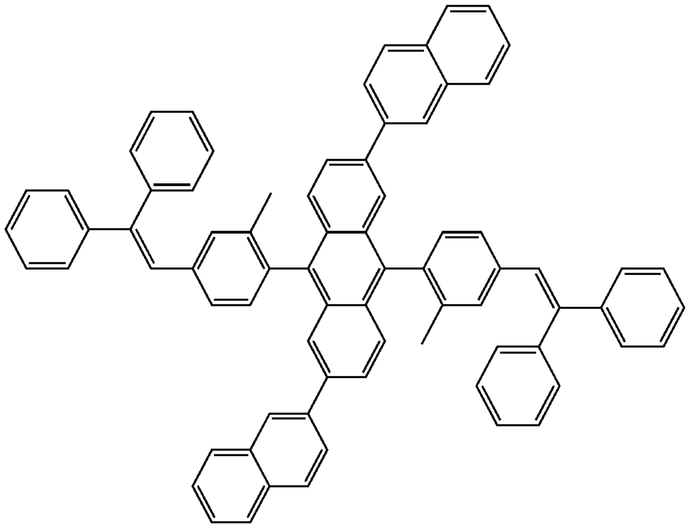

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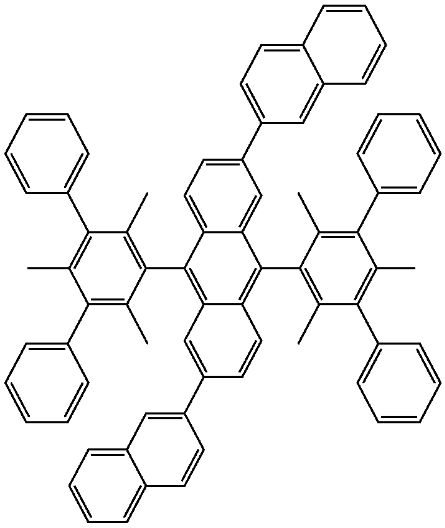

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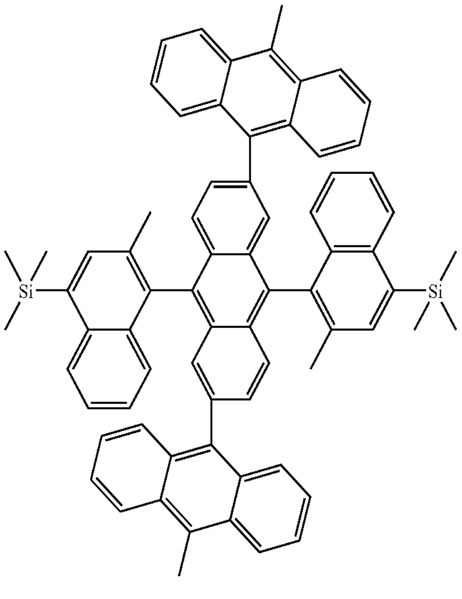

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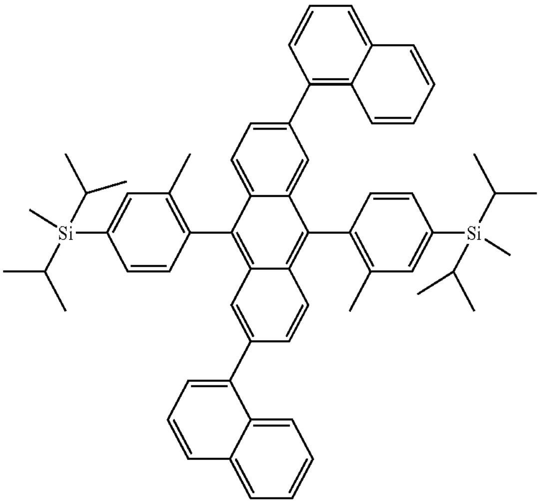

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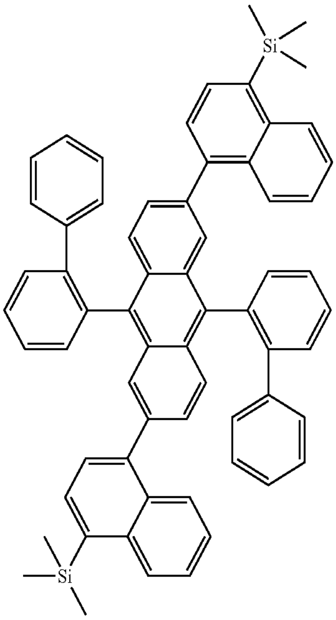

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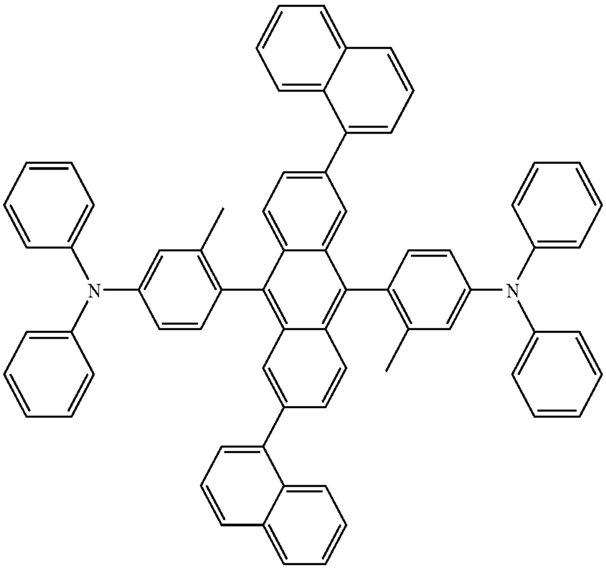

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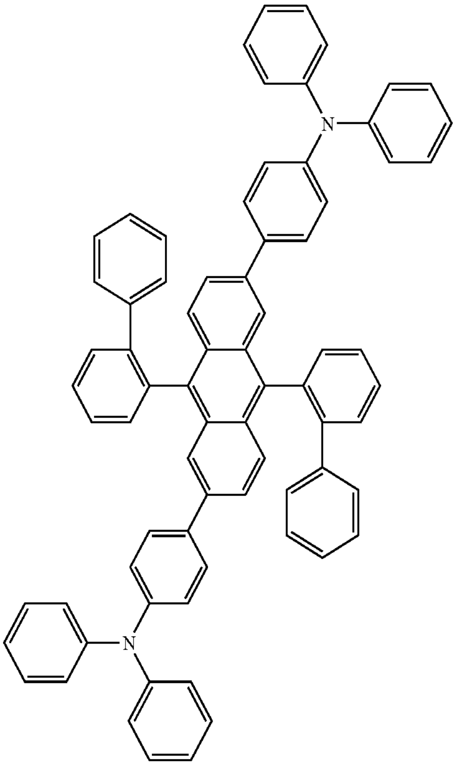

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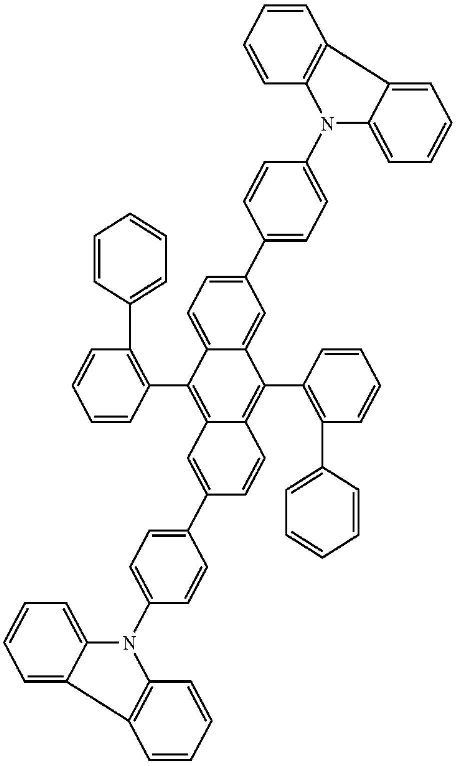

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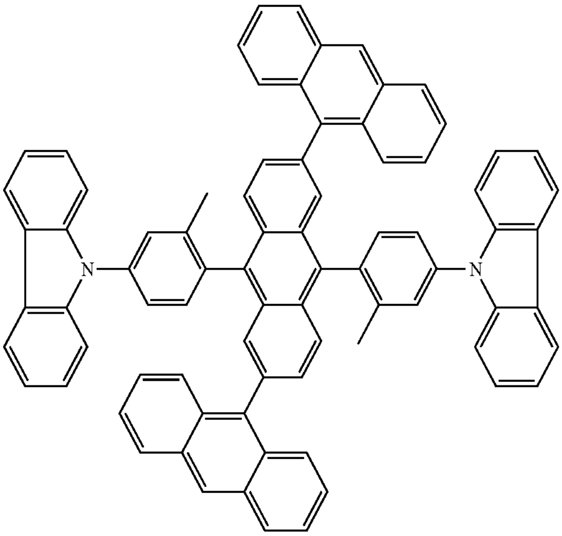

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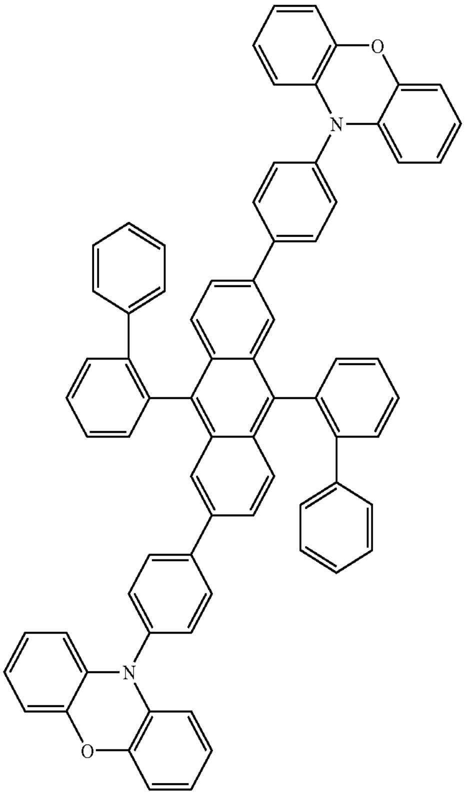

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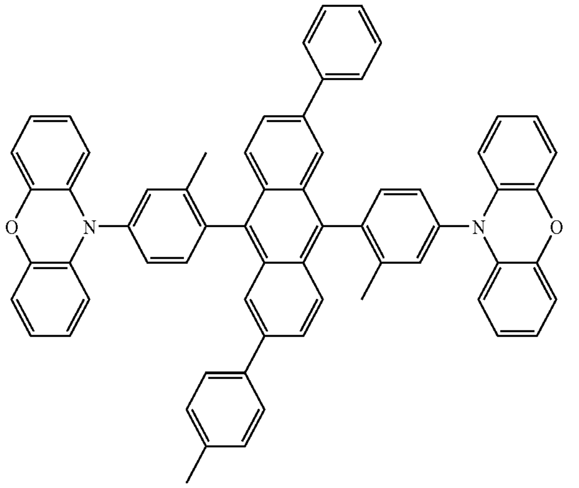

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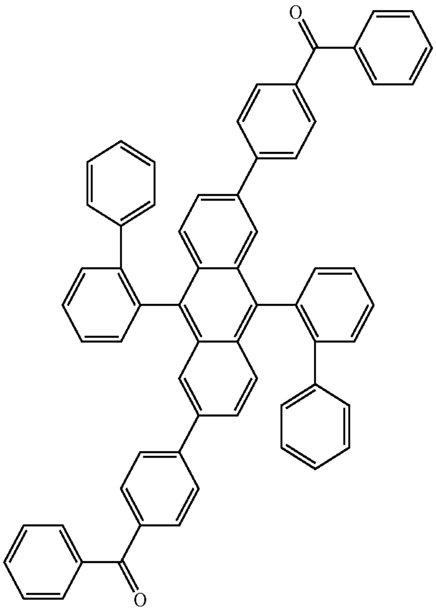

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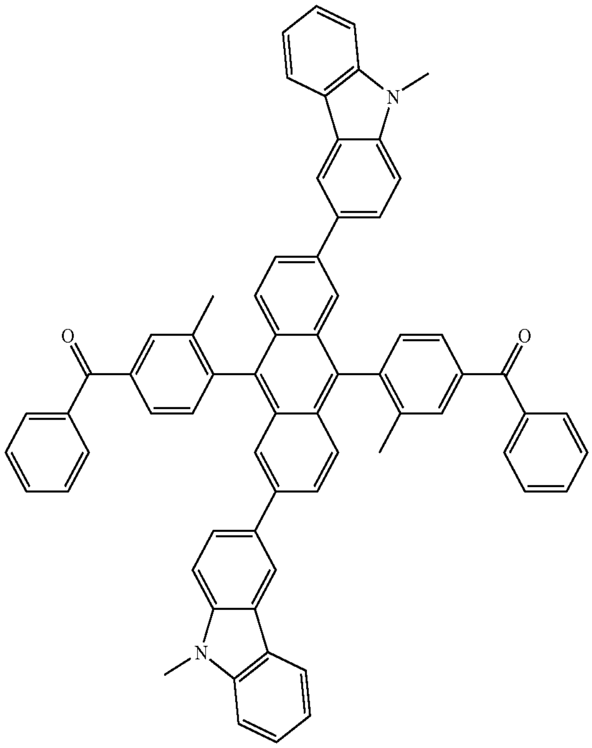

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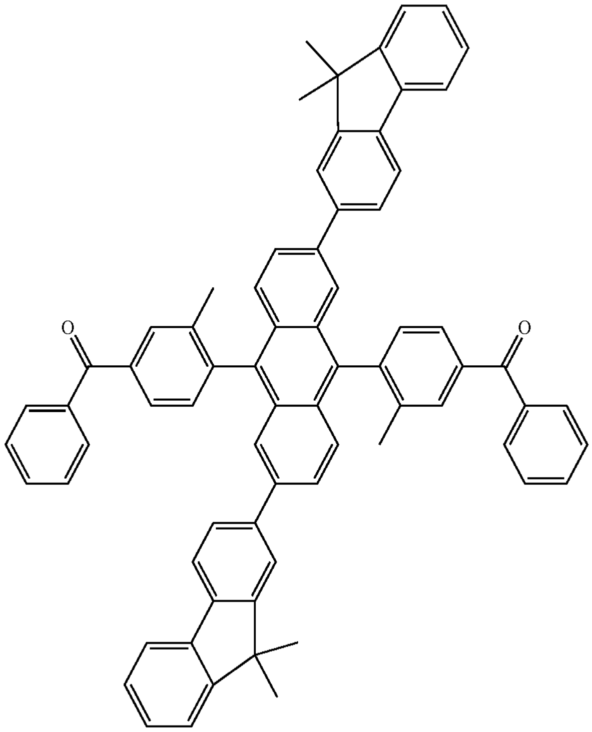

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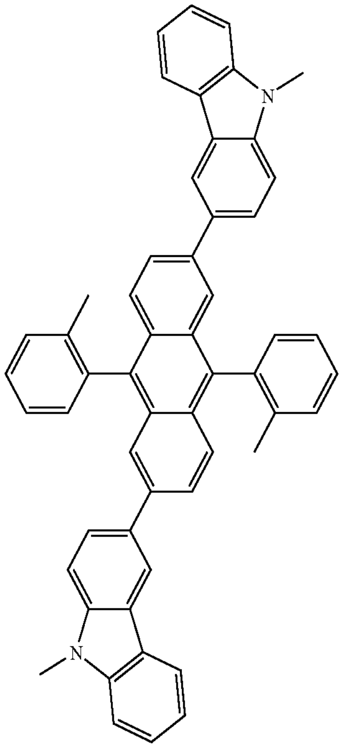

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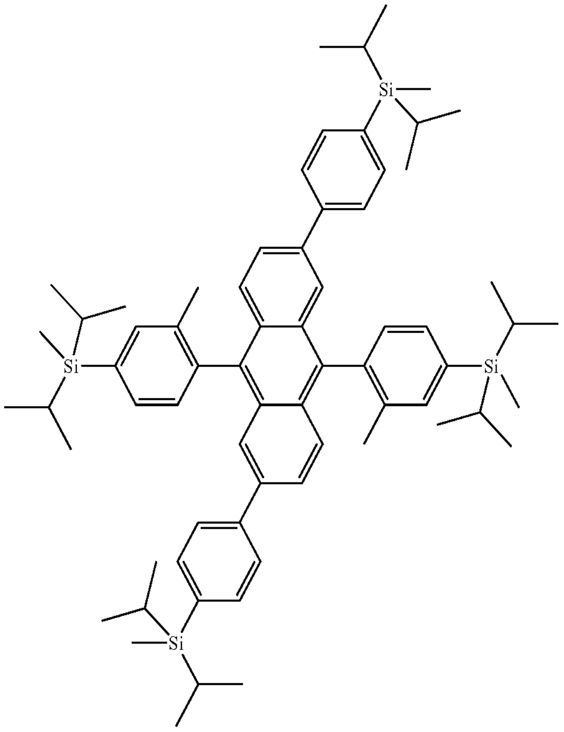

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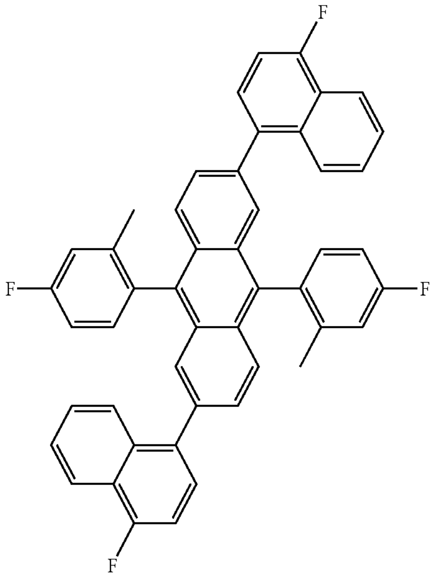

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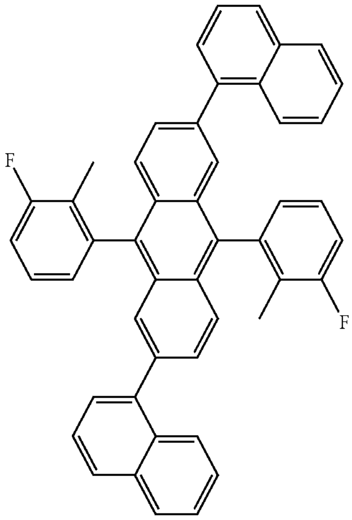

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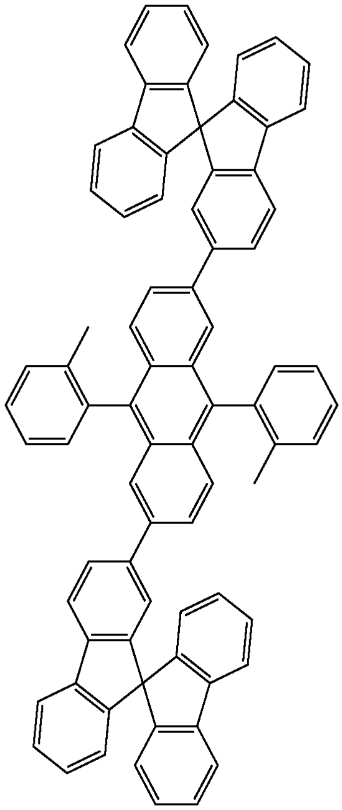

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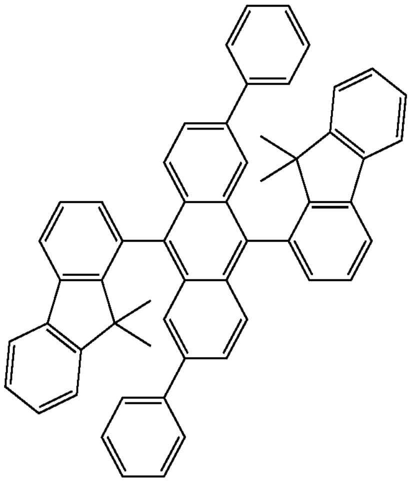

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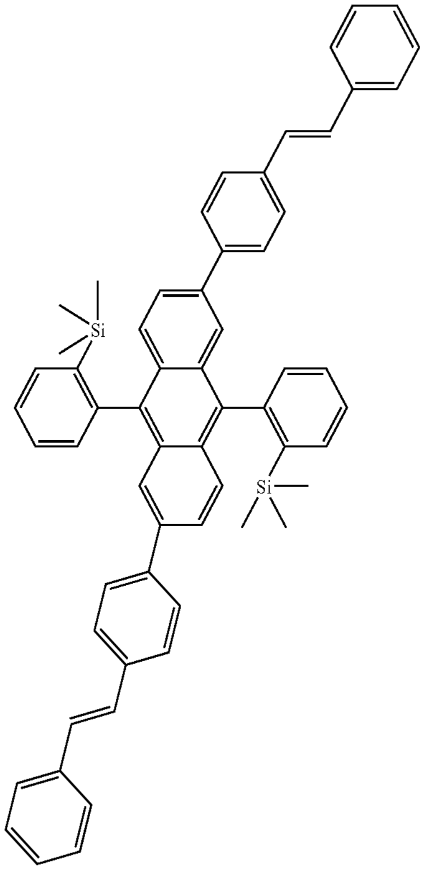

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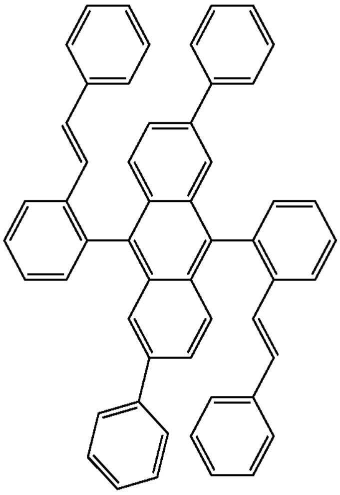

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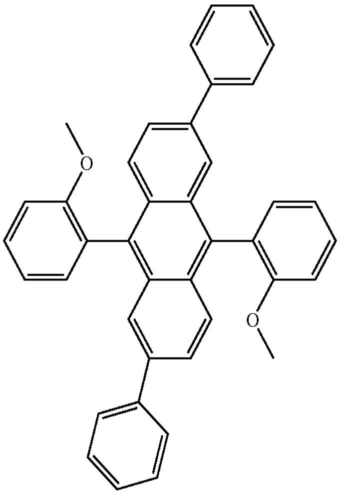

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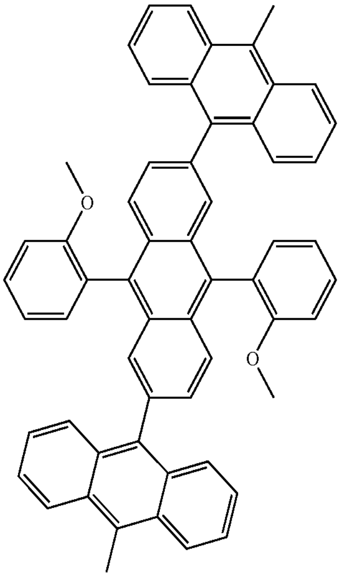

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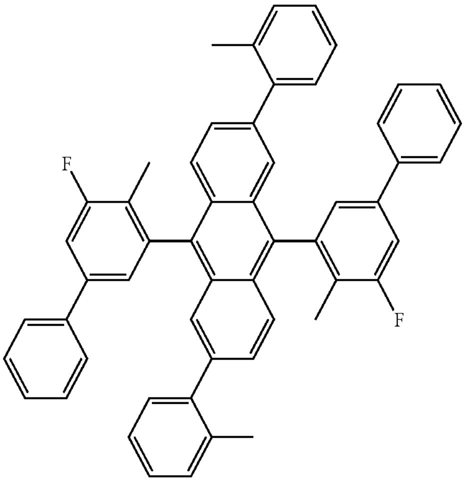

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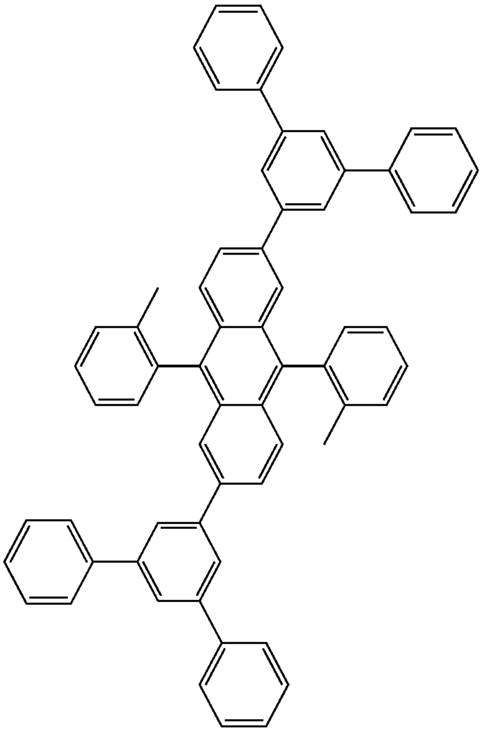

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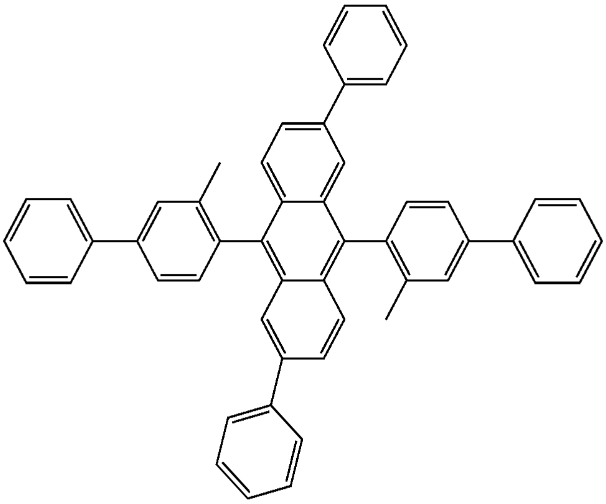

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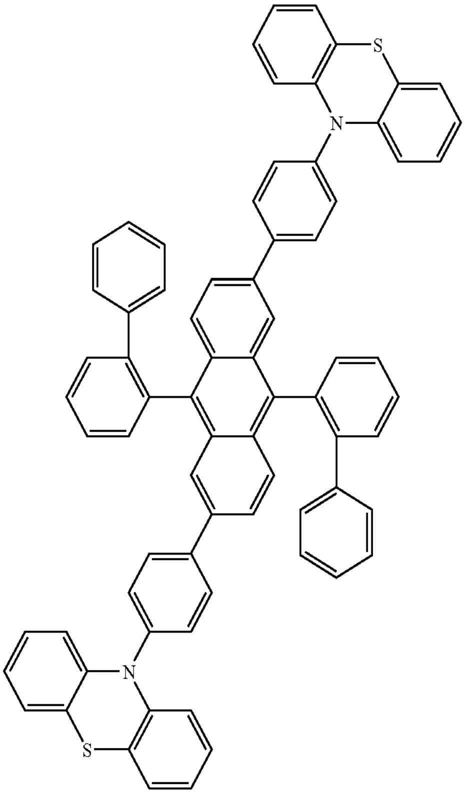

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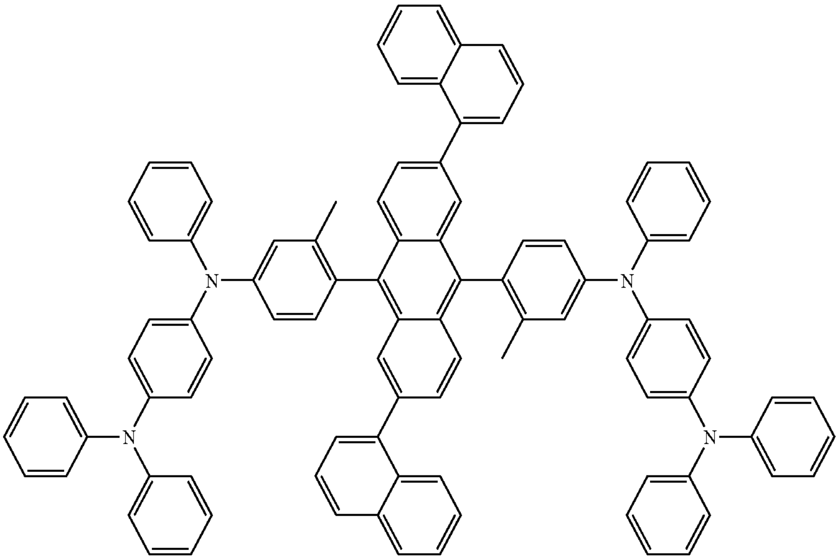

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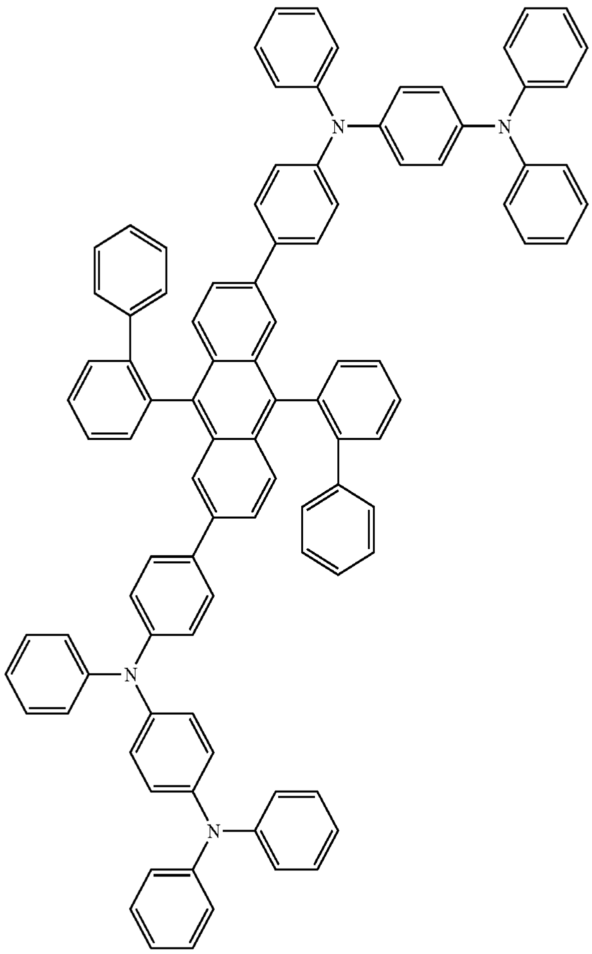
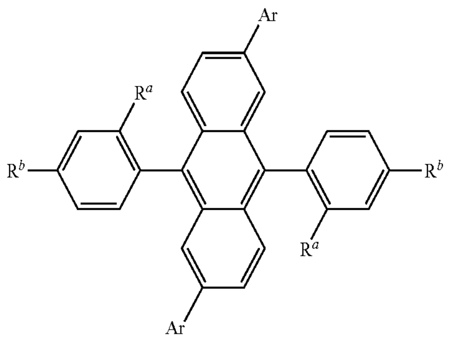


(50)

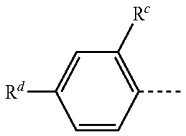
Table 1 below indicates further preferred structures of the formula (1). The symbols and indices used in the table relate to formula (8) depicted below:

Formula (8)



Ar here stands for a group of the formula (9), (10) or (11):

Formula (9)



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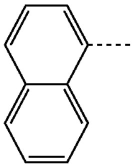
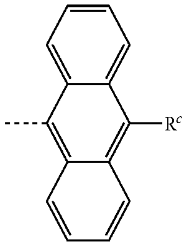
Formula (10)



Formula (11)



where the dashed bond denotes the link to the anthracene unit.

Furthermore, the abbreviation N(p-Tol)$_2$ in Table 1 stands for a bis(para-tolyl)amino group.

TABLE 1

Preferred structures of the formula (8)

		Ar			
No.	Ar	Rc	Rd	Ra	Rb
1	Phenyl	H	H	Methyl	H
2	Phenyl	H	H	Methyl	Methyl
3	Phenyl	H	H	Methyl	tert-Butyl
4	Phenyl	H	H	Methyl	Si(Me)$_3$
5	Phenyl	H	H	Methyl	N(p-Tol)$_2$
6	Phenyl	H	H	Methyl	Phenyl
7	Phenyl	H	H	tert-Butyl	H
8	Phenyl	H	H	tert-Butyl	Methyl
9	Phenyl	H	H	tert-Butyl	tert-Butyl
10	Phenyl	H	H	tert-Butyl	Si(Me)$_3$
11	Phenyl	H	H	tert-Butyl	N(p-Tol)$_2$
12	Phenyl	H	H	tert-Butyl	Phenyl
13	Phenyl	H	H	Si(Me)$_3$	H
14	Phenyl	H	H	Si(Me)$_3$	Methyl
15	Phenyl	H	H	Si(Me)$_3$	tert-Butyl
16	Phenyl	H	H	Si(Me)$_3$	Si(Me)$_3$
17	Phenyl	H	H	Si(Me)$_3$	N(p-Tol)$_2$
18	Phenyl	H	H	Si(Me)$_3$	Phenyl
19	Phenyl	H	H	N(p-Tol)$_2$	H
20	Phenyl	H	H	N(p-Tol)$_2$	Methyl
21	Phenyl	H	H	N(p-Tol)$_2$	tert-Butyl
22	Phenyl	H	H	N(p-Tol)$_2$	Si(Me)$_3$
23	Phenyl	H	H	N(p-Tol)$_2$	N(p-Tol)$_2$
24	Phenyl	H	H	N(p-Tol)$_2$	Phenyl
25	Phenyl	H	H	Phenyl	H
26	Phenyl	H	H	Phenyl	Methyl
27	Phenyl	H	H	Phenyl	tert-Butyl
28	Phenyl	H	H	Phenyl	Si(Me)$_3$
29	Phenyl	H	H	Phenyl	N(p-Tol)$_2$
30	Phenyl	H	H	Phenyl	Phenyl
31	Phenyl	H	Methyl	Methyl	H
32	Phenyl	H	Methyl	Methyl	Methyl
33	Phenyl	H	Methyl	Methyl	tert-Butyl
34	Phenyl	H	Methyl	Methyl	Si(Me)$_3$
35	Phenyl	H	Methyl	Methyl	N(p-Tol)$_2$
36	Phenyl	H	Methyl	Methyl	Phenyl
37	Phenyl	H	Methyl	tert-Butyl	H
38	Phenyl	H	Methyl	tert-Butyl	Methyl
39	Phenyl	H	Methyl	tert-Butyl	tert-Butyl
40	Phenyl	H	Methyl	tert-Butyl	Si(Me)$_3$
41	Phenyl	H	Methyl	tert-Butyl	N(p-Tol)$_2$
42	Phenyl	H	Methyl	tert-Butyl	Phenyl
43	Phenyl	H	Methyl	Si(Me)$_3$	H
44	Phenyl	H	Methyl	Si(Me)$_3$	Methyl
45	Phenyl	H	Methyl	Si(Me)$_3$	tert-Butyl
46	Phenyl	H	Methyl	Si(Me)$_3$	Si(Me)$_3$
47	Phenyl	H	Methyl	Si(Me)$_3$	N(p-Tol)$_2$
48	Phenyl	H	Methyl	Si(Me)$_3$	Phenyl
49	Phenyl	H	Methyl	N(p-Tol)$_2$	H
50	Phenyl	H	Methyl	N(p-Tol)$_2$	Methyl
51	Phenyl	H	Methyl	N(p-Tol)$_2$	tert-Butyl
52	Phenyl	H	Methyl	N(p-Tol)$_2$	Si(Me)$_3$
53	Phenyl	H	Methyl	N(p-Tol)$_2$	N(p-Tol)$_2$
54	Phenyl	H	Methyl	N(p-Tol)$_2$	Phenyl
55	Phenyl	H	Methyl	Phenyl	H
56	Phenyl	H	Methyl	Phenyl	Methyl
57	Phenyl	H	Methyl	Phenyl	tert-Butyl
58	Phenyl	H	Methyl	Phenyl	Si(Me)$_3$
59	Phenyl	H	Methyl	Phenyl	N(p-Tol)$_2$
60	Phenyl	H	Methyl	Phenyl	Phenyl
61	Phenyl	H	tert-Butyl	Methyl	H
62	Phenyl	H	tert-Butyl	Methyl	Methyl
63	Phenyl	H	tert-Butyl	Methyl	tert-Butyl
64	Phenyl	H	tert-Butyl	Methyl	Si(Me)3
65	Phenyl	H	tert-Butyl	Methyl	N(p-Tol)$_2$
66	Phenyl	H	tert-Butyl	Methyl	Phenyl
67	Phenyl	H	tert-Butyl	tert-Butyl	H
68	Phenyl	H	tert-Butyl	tert-Butyl	Methyl
69	Phenyl	H	tert-Butyl	tert-Butyl	tert-Butyl
70	Phenyl	H	tert-Butyl	tert-Butyl	Si(Me)$_3$
71	Phenyl	H	tert-Butyl	tert-Butyl	N(p-Tol)$_2$
72	Phenyl	H	tert-Butyl	tert-Butyl	Phenyl
73	Phenyl	H	tert-Butyl	Si(Me)$_3$	H
74	Phenyl	H	tert-Butyl	Si(Me)$_3$	Methyl
75	Phenyl	H	tert-Butyl	Si(Me)$_3$	tert-Butyl
76	Phenyl	H	tert-Butyl	Si(Me)$_3$	Si(Me)$_3$
77	Phenyl	H	tert-Butyl	Si(Me)$_3$	N(p-Tol)$_2$
78	Phenyl	H	tert-Butyl	Si(Me)$_3$	Phenyl
79	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	H
80	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	Methyl
81	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	tert-Butyl
82	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	Si(Me)$_3$
83	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	N(p-Tol)$_2$
84	Phenyl	H	tert-Butyl	N(p-Tol)$_2$	Phenyl
85	Phenyl	H	tert-Butyl	Phenyl	H
86	Phenyl	H	tert-Butyl	Phenyl	Methyl
87	Phenyl	H	tert-Butyl	Phenyl	tert-Butyl
88	Phenyl	H	tert-Butyl	Phenyl	Si(Me)$_3$
89	Phenyl	H	tert-Butyl	Phenyl	N(p-Tol)$_2$
90	Phenyl	H	tert-Butyl	Phenyl	Phenyl
91	Phenyl	H	Si(Me)$_3$	Methyl	H
92	Phenyl	H	Si(Me)$_3$	Methyl	Methyl
93	Phenyl	H	Si(Me)$_3$	Methyl	tert-Butyl
94	Phenyl	H	Si(Me)$_3$	Methyl	Si(Me)$_3$
95	Phenyl	H	Si(Me)$_3$	Methyl	N(p-Tol)$_2$
96	Phenyl	H	Si(Me)$_3$	Methyl	Phenyl
97	Phenyl	H	Si(Me)$_3$	tert-Butyl	H
98	Phenyl	H	Si(Me)$_3$	tert-Butyl	Methyl
99	Phenyl	H	Si(Me)$_3$	tert-Butyl	tert-Butyl
100	Phenyl	H	Si(Me)$_3$	tert-Butyl	Si(Me)$_3$
101	Phenyl	H	Si(Me)$_3$	tert-Butyl	N(p-Tol)$_2$
102	Phenyl	H	Si(Me)$_3$	tert-Butyl	Phenyl
103	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	H
104	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	Methyl
105	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	tert-Butyl
106	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	Si(Me)$_3$
107	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	N(p-Tol)$_2$
108	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$	Phenyl
109	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	H
110	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	Methyl
111	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	tert-Butyl
112	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	Si(Me)$_3$
113	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	N(p-Tol)$_2$
114	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$	Phenyl
115	Phenyl	H	Si(Me)$_3$	Phenyl	H
116	Phenyl	H	Si(Me)$_3$	Phenyl	Methyl
117	Phenyl	H	Si(Me)$_3$	Phenyl	tert-Butyl
118	Phenyl	H	Si(Me)3	Phenyl	Si(Me)$_3$
119	Phenyl	H	Si(Me)$_3$	Phenyl	N(p-Tol)$_2$
120	Phenyl	H	Si(Me)$_3$	Phenyl	Phenyl
121	Phenyl	H	N(p-Tol)$_2$	Methyl	H
122	Phenyl	H	N(p-Tol)$_2$	Methyl	Methyl
123	Phenyl	H	N(p-Tol)$_2$	Methyl	tert-Butyl
124	Phenyl	H	N(p-Tol)$_2$	Methyl	Si(Me)$_3$
125	Phenyl	H	N(p-Tol)$_2$	Methyl	N(p-Tol)$_2$
126	Phenyl	H	N(p-Tol)$_2$	Methyl	Phenyl
127	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	H
128	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	Methyl
129	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	tert-Butyl
130	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	Si(Me)$_3$
131	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	N(p-Tol)$_2$
132	Phenyl	H	N(p-Tol)$_2$	tert-Butyl	Phenyl
133	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$	H
134	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$	Methyl
135	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$	tert-Butyl
136	Phenyl	H	N(p-Tol)$_2$	Si(Me}$_3$	Si(Me)$_3$
137	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$	N(p-Tol)$_2$
138	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$	Phenyl
139	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	H
140	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	Methyl
141	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	tert-Butyl
142	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	Si(Me)$_3$
143	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	N(p-Tol)$_2$
144	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$	Phenyl

TABLE 1-continued

Preferred structures of the formula (8)

		Ar			
No.	Ar	Rc	Rd	Ra	Rb
145	Phenyl	H	N(p-Tol)$_2$	Phenyl	H
146	Phenyl	H	N(p-Tol)$_2$	Phenyl	Methyl
147	Phenyl	H	N(p-Tol)$_2$	Phenyl	tert-Butyl
148	Phenyl	H	N(p-Tol)$_2$	Phenyl	Si(Me)$_3$
149	Phenyl	H	N(p-Tol)$_2$	Phenyl	N(p-Tol)$_2$
150	Phenyl	H	N(p-Tol)$_2$	Phenyl	Phenyl
151	Phenyl	Methyl	H	Methyl	H
152	Phenyl	Methyl	H	Methyl	Methyl
153	Phenyl	Methyl	H	Methyl	tert-Butyl
154	Phenyl	Methyl	H	Methyl	Si(Me)$_3$
155	Phenyl	Methyl	H	Methyl	N(p-Tol)$_2$
156	Phenyl	Methyl	H	Methyl	Phenyl
157	Phenyl	Methyl	H	tert-Butyl	H
158	Phenyl	Methyl	H	tert-Butyl	Methyl
159	Phenyl	Methyl	H	tert-Butyl	tert-Butyl
160	Phenyl	Methyl	H	tert-Butyl	Si(Me)$_3$
161	Phenyl	Methyl	H	tert-Butyl	N(p-Tol)$_2$
162	Phenyl	Methyl	H	tert-Butyl	Phenyl
163	Phenyl	Methyl	H	Si(Me)$_3$	H
164	Phenyl	Methyl	H	Si(Me)$_3$	Methyl
165	Phenyl	Methyl	H	Si(Me)$_3$	tert-Butyl
166	Phenyl	Methyl	H	Si(Me)$_3$	Si(Me)$_3$
167	Phenyl	Methyl	H	Si(Me)$_3$	N(p-Tol)$_2$
168	Phenyl	Methyl	H	Si(Me)$_3$	Phenyl
169	Phenyl	Methyl	H	N(p-Tol)$_2$	H
170	Phenyl	Methyl	H	N(p-Tol)$_2$	Methyl
171	Phenyl	Methyl	H	N(p-Tol)$_2$	tert-Butyl
172	Phenyl	Methyl	H	N(p-Tol)$_2$	Si(Me)$_3$
173	Phenyl	Methyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$
174	Phenyl	Methyl	H	N(p-Tol)$_2$	Phenyl
175	Phenyl	Methyl	H	Phenyl	H
176	Phenyl	Methyl	H	Phenyl	Methyl
177	Phenyl	Methyl	H	Phenyl	tert-Butyl
178	Phenyl	Methyl	H	Phenyl	Si(Me)$_3$
179	Phenyl	Methyl	H	Phenyl	N(p-Tol)$_2$
180	Phenyl	Methyl	H	Phenyl	Phenyl
181	Phenyl	tert-Butyl	H	Methyl	H
182	Phenyl	tert-Butyl	H	Methyl	Methyl
183	Phenyl	tert-Butyl	H	Methyl	tert-Butyl
184	Phenyl	tert-Butyl	H	Methyl	Si(Me)$_3$
185	Phenyl	tert-Butyl	H	Methyl	N(p-Tol)$_2$
186	Phenyl	tert-Butyl	H	Methyl	Phenyl
187	Phenyl	tert-Butyl	H	tert-Butyl	H
188	Phenyl	tert-Butyl	H	tert-Butyl	Methyl
189	Phenyl	tert-Butyl	H	tert-Butyl	tert-Butyl
190	Phenyl	tert-Butyl	H	tert-Butyl	Si(Me)$_3$
191	Phenyl	tert-Butyl	H	tert-Butyl	N(p-Tol)$_2$
192	Phenyl	tert-Butyl	H	tert-Butyl	Phenyl
193	Phenyl	tert-Butyl	H	Si(Me)$_3$	H
194	Phenyl	tert-Butyl	H	Si(Me)$_3$	Methyl
195	Phenyl	tert-Butyl	H	Si(Me)$_3$	tert-Butyl
196	Phenyl	tert-Butyl	H	Si(Me)$_3$	Si(Me)$_3$
197	Phenyl	tert-Butyl	H	Si(Me)3	N(p-Tol)$_2$
198	Phenyl	tert-Butyl	H	Si(Me)$_3$	Phenyl
199	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	H
200	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	Methyl
201	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	tert-Butyl
202	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	Si(Me)$_3$
203	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$
204	Phenyl	tert-Butyl	H	N(p-Tol)$_2$	Phenyl
205	Phenyl	tert-Butyl	H	Phenyl	H
206	Phenyl	tert-Butyl	H	Phenyl	Methyl
207	Phenyl	tert-Butyl	H	Phenyl	tert-Butyl
208	Phenyl	tert-Butyl	H	Phenyl	Si(Me)$_3$
209	Phenyl	tert-Butyl	H	Phenyl	N(p-Tol)$_2$
210	Phenyl	tert-Butyl	H	Phenyl	Phenyl
211	Phenyl	Si(Me)$_3$	H	Methyl	H
212	Phenyl	Si(Me)$_3$	H	Methyl	Methyl
213	Phenyl	Si(Me)$_3$	H	Methyl	tert-Butyl
214	Phenyl	Si(Me)$_3$	H	Methyl	Si(Me)$_3$
215	Phenyl	Si(Me)$_3$	H	Methyl	N(p-Tol)$_2$
216	Phenyl	Si(Me)$_3$	H	Methyl	Phenyl
217	Phenyl	Si(Me)$_3$	H	tert-Butyl	H
218	Phenyl	Si(Me)$_3$	H	tert-Butyl	Methyl

TABLE 1-continued

Preferred structures of the formula (8)

		Ar			
No.	Ar	Rc	Rd	Ra	Rb
219	Phenyl	Si(Me)$_3$	H	tert-Butyl	tert-Butyl
220	Phenyl	Si(Me)$_3$	H	tert-Butyl	Si(Me)$_3$
221	Phenyl	Si(Me)$_3$	H	tert-Butyl	N(p-Tol)$_2$
222	Phenyl	Si(Me)$_3$	H	tert-Butyl	Phenyl
223	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	H
224	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	Methyl
225	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	tert-Butyl
226	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	Si(Me)$_3$
227	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	N(p-Tol)$_2$
228	Phenyl	Si(Me)3	H	Si(Me)3	Phenyl
229	Phenyl	Si(Me)$_3$	H	Si(Me)$_3$	H
230	Phenyl	Si(Me)$_3$	H	N(p-Tol)$_2$	Methyl
231	Phenyl	Si(Me)$_3$	H	N(p-Tol)$_2$	tert-Butyl
232	Phenyl	Si(Me)$_3$	H	N(p-Tol)$_2$	Si(Me)$_3$
233	Phenyl	Si(Me)$_3$	H	N(p-Tol)$_2$	N(p-Tol)$_2$
234	Phenyl	Si(Me)$_3$	H	N(p-Tol)$_2$	Phenyl
235	Phenyl	Si(Me)$_3$	H	Phenyl	H
236	Phenyl	Si(Me)$_3$	H	Phenyl	Methyl
237	Phenyl	Si(Me)$_3$	H	Phenyl	tert-Butyl
238	Phenyl	Si(Me)$_3$	H	Phenyl	Si(Me)$_3$
239	Phenyl	Si(Me)$_3$	H	Phenyl	N(p-Tol)$_2$
240	Phenyl	Si(Me)$_3$	H	Phenyl	Phenyl
241	Phenyl	N(p-Tol)$_2$	H	Methyl	H
242	Phenyl	N(p-Tol)$_2$	H	Methyl	Methyl
243	Phenyl	N(p-Tol)$_2$	H	Methyl	tert-Butyl
244	Phenyl	N(p-Tol)$_2$	H	Methyl	Si(Me)$_3$
245	Phenyl	N(p-Tol)$_2$	H	Methyl	N(p-Tol)$_2$
246	Phenyl	N(p-Tol)$_2$	H	Methyl	Phenyl
247	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	H
248	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	Methyl
249	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	tert-Butyl
250	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	Si(Me)$_3$
251	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	N(p-Tol)$_2$
252	Phenyl	N(p-Tol)$_2$	H	tert-Butyl	Phenyl
253	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	H
254	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	Methyl
255	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	tert-Butyl
256	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	Si(Me)$_3$
257	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	N(p-Tol)$_2$
258	Phenyl	N(p-Tol)$_2$	H	Si(Me)$_3$	Phenyl
259	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	H
260	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	Methyl
261	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	tert-Butyl
262	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	Si(Me)$_3$
263	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	N(p-Tol)$_2$
264	Phenyl	N(p-Tol)$_2$	H	N(p-Tol)$_2$	Phenyl
265	Phenyl	N(p-Tol)$_2$	H	Phenyl	H
266	Phenyl	N(p-Tol)$_2$	H	Phenyl	Methyl
267	Phenyl	N(p-Tol)$_2$	H	Phenyl	tert-Butyl
268	Phenyl	N(p-Tol)$_2$	H	Phenyl	Si(Me)$_3$
269	Phenyl	N(p-Tol)$_2$	H	Phenyl	N(p-Tol)$_2$
270	Phenyl	N(p-Tol)$_2$	H	Phenyl	Phenyl
271	Phenyl	Phenyl	H	Methyl	H
272	Phenyl	Phenyl	H	Methyl	Methyl
273	Phenyl	Phenyl	H	Methyl	tert-Butyl
274	Phenyl	Phenyl	H	Methyl	Si(Me)$_3$
275	Phenyl	Phenyl	H	Methyl	N(p-Tol)$_2$
276	Phenyl	Phenyl	H	Methyl	Phenyl
277	Phenyl	Phenyl	H	tert-Butyl	H
278	Phenyl	Phenyl	H	tert-Butyl	Methyl
279	Phenyl	Phenyl	H	tert-Butyl	tert-Butyl
280	Phenyl	Phenyl	H	tert-Butyl	Si(Me)$_3$
281	Phenyl	Phenyl	H	tert-Butyl	N(p-Tol)$_2$
282	Phenyl	Phenyl	H	tert-Butyl	Phenyl
283	Phenyl	Phenyl	H	Si(Me)$_3$	H
284	Phenyl	Phenyl	H	Si(Me)$_3$	Methyl
285	Phenyl	Phenyl	H	Si(Me)$_3$	tert-Butyl
286	Phenyl	Phenyl	H	Si(Me)$_3$	Si(Me)$_3$
287	Phenyl	Phenyl	H	Si(Me)$_3$	N(p-Tol)$_2$
288	Phenyl	Phenyl	H	Si(Me)$_3$	Phenyl
289	Phenyl	Phenyl	H	N(p-Tol)$_2$	H
290	Phenyl	Phenyl	H	N(p-Tol)$_2$	Methyl
291	Phenyl	Phenyl	H	N(p-Tol)$_2$	tert-Butyl
292	Phenyl	Phenyl	H	N(p-Tol)$_2$	Si(Me)$_3$

TABLE 1-continued

Preferred structures of the formula (8)

		Ar			
No.	Ar	Rc	Rd	Ra	Rb
293	Phenyl	Phenyl	H	N(p-Tol)$_2$	N(p-Tol)$_2$
294	Phenyl	Phenyl	H	N(p-Tol)$_2$	Phenyl
295	Phenyl	Phenyl	H	Phenyl	H
296	Phenyl	Phenyl	H	Phenyl	Methyl
297	Phenyl	Phenyl	H	Phenyl	tert-Butyl
298	Phenyl	Phenyl	H	Phenyl	Si(Me)$_3$
299	Phenyl	Phenyl	H	Phenyl	N(p-Tol)$_2$
300	Phenyl	Phenyl	H	Phenyl	Phenyl
301	1-Naphthyl	H	—	Methyl	H
302	1-Naphthyl	H	—	Methyl	Methyl
303	1-Naphthyl	H	—	Methyl	tert-Butyl
304	1-Naphthyl	H	—	Methyl	Si(Me)$_3$
305	1-Naphthyl	H	—	Methyl	N(p-Tol)$_2$
306	1-Naphthyl	H	—	Methyl	Phenyl
307	1-Naphthyl	H	—	tert-Butyl	H
308	1-Naphthyl	H	—	tert-Butyl	Methyl
309	1-Naphthyl	H	—	tert-Butyl	tert-Butyl
310	1-Naphthyl	H	—	tert-Butyl	Si(Me)$_3$
311	1-Naphthyl	H	—	tert-Butyl	N(p-Tol)$_2$
312	1-Naphthyl	H	—	tert-Butyl	Phenyl
313	1-Naphthyl	H	—	Si(Me)$_3$	H
314	1-Naphthyl	H	—	Si(Me)$_3$	Methyl
315	1-Naphthyl	H	—	Si(Me)$_3$	tert-Butyl
316	1-Naphthyl	H	—	Si(Me)$_3$	Si(Me)$_3$
317	1-Naphthyl	H	—	Si(Me)$_3$	N(p-Tol)2
318	1-Naphthyl	H	—	Si(Me)$_3$	N(p-Tol)$_2$
319	1-Naphthyl	H	—	N(p-Tol)$_2$	H
320	1-Naphthyl	H	—	N(p-Tol)$_2$	Methyl
321	1-Naphthyl	H	—	N(p-Tol)$_2$	tert-Butyl
322	1-Naphthyl	H	—	N(p-Tol)$_2$	Si(Me)$_3$
323	1-Naphthyl	H	—	N(p-Tol)$_2$	N(p-Tol)$_2$
324	1-Naphthyl	H	—	N(p-Tol)$_2$	Phenyl
325	1-Naphthyl	H	—	Phenyl	H
326	1-Naphthyl	H	—	Phenyl	Methyl
327	1-Naphthyl	H	—	Phenyl	tert-Butyl
328	1-Naphthyl	H	—	Phenyl	Si(Me)$_3$
329	1-Naphthyl	H	—	Phenyl	N(p-Tol)$_2$
330	1-Naphthyl	H	—	Phenyl	Phenyl
331	1-Naphthyl	Methyl	—	Methyl	H
332	1-Naphthyl	Methyl	—	Methyl	Methyl
333	1-Naphthyl	Methyl	—	Methyl	tert-Butyl
334	1-Naphthyl	Methyl	—	Methyl	Si(Me)$_3$
335	1-Naphthyl	Methyl	—	Methyl	N(p-Tol)$_2$
336	1-Naphthyl	Methyl	—	Methyl	Phenyl
337	1-Naphthyl	Methyl	—	tert-Butyl	H
338	1-Naphthyl	Methyl	—	tert-Butyl	Methyl
339	1-Naphthyl	Methyl	—	tert-Butyl	tert-Butyl
340	1-Naphthyl	Methyl	—	tert-Butyl	Si(Me)$_3$
341	1-Naphthyl	Methyl	—	tert-Butyl	N(p-Tol)$_2$
342	1-Naphthyl	Methyl	—	tert-Butyl	Phenyl
343	1-Naphthyl	Methyl	—	Si(Me)$_3$	H
344	1-Naphthyl	Methyl	—	Si(Me)$_3$	Methyl
345	1-Naphthyl	Methyl	—	Si(Me)$_3$	tert-Butyl
346	1-Naphthyl	Methyl	—	Si(Me)$_3$	Si(Me)$_3$
347	1-Naphthyl	Methyl	—	Si(Me)$_3$	N(p-Tol)$_2$
348	1-Naphthyl	Methyl	—	Si(Me)$_3$	Phenyl
349	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	H
350	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	Methyl
351	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	tert-Butyl
352	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	Si(Me)$_3$
353	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	N(p-Tol)$_2$
354	1-Naphthyl	Methyl	—	N(p-Tol)$_2$	Phenyl
355	1-Naphthyl	Methyl	—	Phenyl	H
356	1-Naphthyl	Methyl	—	Phenyl	Methyl
357	1-Naphthyl	Methyl	—	Phenyl	tert-Butyl
358	1-Naphthyl	Methyl	—	Phenyl	Si(Me)$_3$
359	1-Naphthyl	Methyl	—	Phenyl	N(p-Tol)$_2$
360	1-Naphthyl	Methyl	—	Phenyl	Phenyl
361	2-Naphthyl	—	—	Methyl	H
362	2-Naphthyl	—	—	Methyl	Methyl
363	2-Naphthyl	—	—	Methyl	tert-Butyl
364	2-Naphthyl	—	—	Methyl	Si(Me)$_3$
365	2-Naphthyl	—	—	Methyl	N(p-Tol)$_2$
366	2-Naphthyl	—	—	Methyl	Phenyl
367	2-Naphthyl	—	—	tert-Butyl	H
368	2-Naphthyl	—	—	tert-Butyl	Methyl
369	2-Naphthyl	—	—	tert-Butyl	tert-Butyl
370	2-Naphthyl	—	—	tert-Butyl	Si(Me)$_3$
371	2-Naphthyl	—	—	tert-Butyl	N(p-Tol)$_2$
372	2-Naphthyl	—	—	tert-Butyl	Phenyl
373	2-Naphthyl	—	—	Si(Me)$_3$	H
374	2-Naphthyl	—	—	Si(Me)$_3$	Methyl
375	2-Naphthyl	—	—	Si(Me)$_3$	tert-Butyl
376	2-Naphthyl	—	—	Si(Me)$_3$	Si(Me)$_3$
377	2-Naphthyl	—	—	Si(Me)$_3$	N(p-Tol)$_2$
378	2-Naphthyl	—	—	Si(Me)$_3$	Phenyl
379	2-Naphthyl	—	—	N(p-Tol)$_2$	H
380	2-Naphthyl	—	—	N(p-Tol)$_2$	Methyl
381	2-Naphthyl	—	—	N(p-Tol)$_2$	tert-Butyl
382	2-Naphthyl	—	—	N(p-Tol)$_2$	Si(Me)$_3$
383	2-Naphthyl	—	—	N(p-Tol)$_2$	N(p-Tol)$_2$
384	2-Naphthyl	—	—	N(p-Tol)$_2$	Phenyl
385	2-Naphthyl	—	—	Phenyl	H
386	2-Naphthyl	—	—	Phenyl	Methyl
387	2-Naphthyl	—	—	Phenyl	tert-Butyl
388	2-Naphthyl	—	—	Phenyl	Si(Me)$_3$
389	2-Naphthyl	—	—	Phenyl	N(p-Tol)$_2$
390	2-Naphthyl	—	—	Phenyl	Phenyl
391	9-Anthryl	1-Naphthyl	—	Methyl	H
392	9-Anthryl	1-Naphthyl	—	Methyl	Methyl
393	9-Anthryl	1-Naphthyl	—	Methyl	tert-Butyl
394	9-Anthryl	1-Naphthyl	—	Methyl	Si(Me)$_3$
395	9-Anthryl	1-Naphthyl	—	Methyl	N(p-Tol)$_2$
396	9-Anthryl	1-Naphthyl	—	Methyl	Phenyl
397	9-Anthryl	1-Naphthyl	—	tert-Butyl	H
398	9-Anthryl	1-Naphthyl	—	tert-Butyl	Methyl
399	9-Anthryl	1-Naphthyl	—	tert-Butyl	tert-Butyl
400	9-Anthryl	1-Naphthyl	—	tert-Butyl	Si(Me)$_3$
401	9-Anthryl	1-Naphthyl	—	tert-Butyl	N(p-Tol)$_2$
402	9-Anthryl	1-Naphthyl	—	tert-Butyl	Phenyl
403	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	H
404	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	Methyl
405	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	tert-Butyl
406	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	Si(Me)$_3$
407	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	N(p-Tol)$_2$
408	9-Anthryl	1-Naphthyl	—	Si(Me)$_3$	Phenyl
409	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	H
410	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	Methyl
411	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	tert-Butyl
412	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	Si(Me)$_3$
413	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	N(p-Tol)$_2$
414	9-Anthryl	1-Naphthyl	—	N(p-Tol)$_2$	Phenyl
415	9-Anthryl	1-Naphthyl	—	Phenyl	H
416	9-Anthryl	1-Naphthyl	—	Phenyl	Methyl
417	9-Anthryl	1-Naphthyl	—	Phenyl	tert-Butyl
418	9-Anthryl	1-Naphthyl	—	Phenyl	Si(Me)$_3$
419	9-Anthryl	1-Naphthyl	—	Phenyl	N(p-Tol)$_2$
420	9-Anthryl	1-Naphthyl	—	Phenyl	Phenyl
421	9-Anthryl	2-Naphthyl	—	Methyl	H
422	9-Anthryl	2-Naphthyl	—	Methyl	Methyl
423	9-Anthryl	2-Naphthyl	—	Methyl	tert-Butyl
424	9-Anthryl	2-Naphthyl	—	Methyl	Si(Me)$_3$
425	9-Anthryl	2-Naphthyl	—	Methyl	N(p-Tol)$_2$
426	9-Anthryl	2-Naphthyl	—	Methyl	Phenyl
427	9-Anthryl	2-Naphthyl	—	tert-Butyl	H
428	9-Anthryl	2-Naphthyl	—	tert-Butyl	Methyl
429	9-Anthryl	2-Naphthyl	—	tert-Butyl	tert-Butyl
430	9-Anthryl	2-Naphthyl	—	tert-Butyl	Si(Me)$_3$
431	9-Anthryl	2-Naphthyl	—	tert-Butyl	N(p-Tol)$_2$
432	9-Anthryl	2-Naphthyl	—	tert-Butyl	Phenyl
433	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	H
434	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	Methyl
435	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	tert-Butyl
436	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	Si(Me)$_3$
437	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	N(p-Tol)$_2$
438	9-Anthryl	2-Naphthyl	—	Si(Me)$_3$	Phenyl
439	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	H
440	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	Methyl

TABLE 1-continued

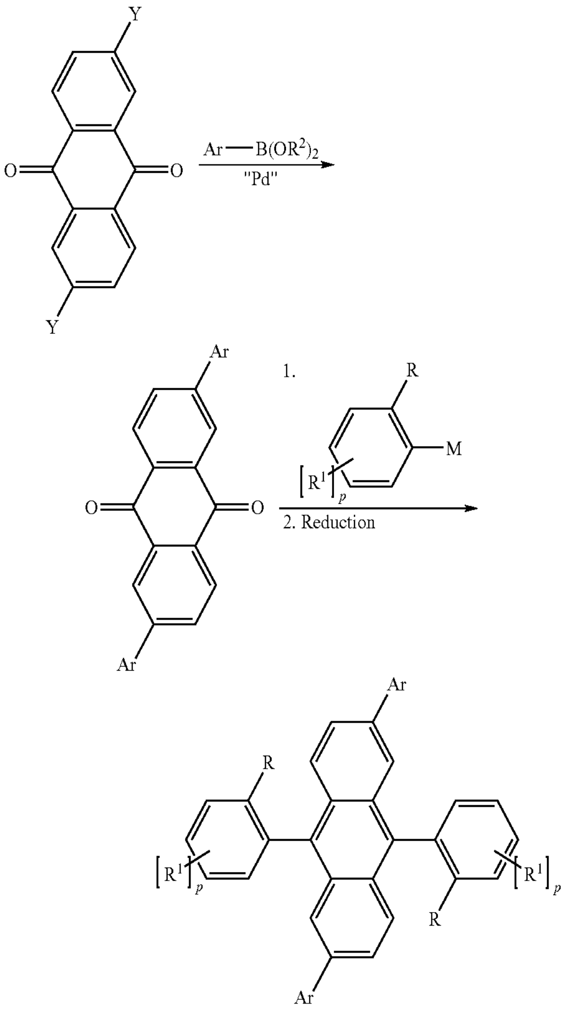
Preferred structures of the formula (8)

		Ar			
No.	Ar	Rc	Rd	Ra	Rb
441	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	tert-Butyl
442	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	Si(Me)$_3$
443	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	N(p-Tol)$_2$
444	9-Anthryl	2-Naphthyl	—	N(p-Tol)$_2$	Phenyl
445	9-Anthryl	2-Naphthyl	—	Phenyl	H
446	9-Anthryl	2-Naphthyl	—	Phenyl	Methyl
447	9-Anthryl	2-Naphthyl	—	Phenyl	tert-Butyl
448	9-Anthryl	2-Naphthyl	—	Phenyl	Si(Me)$_3$
449	9-Anthryl	2-Naphthyl	—	Phenyl	N(p-Tol)$_2$
450	9-Anthryl	2-Naphthyl	—	Phenyl	Phenyl
451	9-Anthryl	N(p-Tol)$_2$	—	Methyl	H
452	9-Anthryl	N(p-Tol)$_2$	—	Methyl	Methyl
453	9-Anthryl	N(p-Tol)$_2$	—	Methyl	tert-Butyl
454	9-Anthryl	N(p-Tol)$_2$	—	Methyl	Si(Me)$_3$
455	9-Anthryl	N(p-Tol)$_2$	—	Methyl	N(p-Tol)$_2$
456	9-Anthryl	N(p-Tol)$_2$	—	Methyl	Phenyl
457	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	H
458	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	Methyl
459	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	tert-Butyl
460	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	Si(Me)$_3$
461	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	N(p-Tol)$_2$
462	9-Anthryl	N(p-Tol)$_2$	—	tert-Butyl	Phenyl
463	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	H
464	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	Methyl
465	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	tert-Butyl
466	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	Si(Me)$_3$
467	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	N(p-Tol)$_2$
468	9-Anthryl	N(p-Tol)$_2$	—	Si(Me)$_3$	Phenyl
469	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	H
470	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	Methyl
471	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	tert-Butyl
472	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	Si(Me)$_3$
473	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	N(p-Tol)$_2$
474	9-Anthryl	N(p-Tol)$_2$	—	N(p-Tol)$_2$	Phenyl
475	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	H
476	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	Methyl
477	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	tert-Butyl
478	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	Si(Me)$_3$
479	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	N(p-Tol)$_2$
480	9-Anthryl	N(p-Tol)$_2$	—	Phenyl	Phenyl
481	9-Anthryl	Phenyl	—	Methyl	H
482	9-Anthryl	Phenyl	—	Methyl	Methyl
483	9-Anthryl	Phenyl	—	Methyl	tert-Butyl
484	9-Anthryl	Phenyl	—	Methyl	Si(Me)$_3$
485	9-Anthryl	Phenyl	—	Methyl	N(p-Tol)$_2$
486	9-Anthryl	Phenyl	—	Methyl	Phenyl
487	9-Anthryl	Phenyl	—	tert-Butyl	H
488	9-Anthryl	Phenyl	—	tert-Butyl	Methyl
489	9-Anthryl	Phenyl	—	tert-Butyl	tert-Butyl
490	9-Anthryl	Phenyl	—	tert-Butyl	Si(Me)$_3$
491	9-Anthryl	Phenyl	—	tert-Butyl	N(p-Tol)$_2$
492	9-Anthryl	Phenyl	—	tert-Butyl	Phenyl
493	9-Anthryl	Phenyl	—	Si(Me)$_3$	H
494	9-Anthryl	Phenyl	—	Si(Me)$_3$	Methyl
495	9-Anthryl	Phenyl	—	Si(Me)$_3$	tert-Butyl
496	9-Anthryl	Phenyl	—	Si(Me)$_3$	Si(Me)$_3$
497	9-Anthryl	Phenyl	—	Si(Me)$_3$	N(p-Tol)$_2$
498	9-Anthryl	Phenyl	—	Si(Me)$_3$	Phenyl
499	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	H
500	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	Methyl
501	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	tert-Butyl
502	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	Si(Me)$_3$
503	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	N(p-Tol)$_2$
504	9-Anthryl	Phenyl	—	N(p-Tol)$_2$	Phenyl
505	9-Anthryl	Phenyl	—	Phenyl	H
506	9-Anthryl	Phenyl	—	Phenyl	Methyl
507	9-Anthryl	Phenyl	—	Phenyl	tert-Butyl
508	9-Anthryl	Phenyl	—	Phenyl	Si(Me)$_3$
509	9-Anthryl	Phenyl	—	Phenyl	N(p-Tol)$_2$
510	9-Anthryl	Phenyl	—	Phenyl	Phenyl

The compounds can be synthesised, for example, starting from 2,6-dichloro- or dibromoanthraquinone. This is reacted with arylboronic acids in a Suzuki coupling to give the corresponding 2,6-diarylanthraquinone. This can be reacted with an aromatic Grignard reagent in a further step and then with a reducing agent, for example tin(II) chloride, to give 2,6,9,10-tetraarylanthracene.

The present invention therefore furthermore relates to a process for the preparation of compounds of the formula (1) by reaction of 2,6-dihaloanthraquinone or an analogous sulfonic acid derivative with a boronic acid derivative of the group Ar with palladium catalysis, followed by reaction with a corresponding ortho-substituted organometallic phenyl derivative and reduction.

The process thus takes place in accordance with the following scheme:



Ar, R, R^1, R^2 and p here have the same meanings as described above. Y stands for chlorine, bromine or iodine, preferably bromine, or for a group of the formula OSO$_2$R^2. M stands for an electropositive metal, in particular lithium, magnesium or zinc, and, in the case of a divalent metal, also contains a further organic group or a group Y. The way in which a Suzuki coupling (first reaction step) is carried out and which palladium catalysts are particularly suitable for this purpose is known to the person skilled in the art of organic synthesis. The reducing agent employed in the second reaction step is preferably tin(II) chloride.

The compounds of the formula (1) can be employed in organic electroluminescent devices. They are particularly suitable for use as host material for fluorescent emitters, but may each of which may, depending on the substitution pattern, also be employed as emitter, as hole-transport material, as hole-blocking material and/or as electron-transport material.

The invention therefore furthermore relates to the use of compounds of the formula (1) in organic electronic devices, in particular in organic electroluminescent devices, in particular as host material, as emitter, as hole-transport material, as hole-blocking material and/or as electron-transport material.

The invention furthermore relates to organic electronic devices, in particular organic electroluminescent devices, comprising anode, cathode and at least one emitting layer, where at least one layer comprises at least one compound of the formula (1). The layer which comprises the compound of the formula (1) is preferably an emitting layer, a hole-transport layer, a hole-injection layer, a hole-blocking layer or an electron-transport layer.

Apart from the cathode, anode and emitting layer (or emitting layers), the organic electroluminescent device may also comprise further layers. These may be, for example: hole-injection layer, hole-transport layer, hole-blocking layer, electron-transport layer, electron-injection layer and/or a charge-generation layer (T. Matsumoto et al., *Multiphoton Organic EL Device Having Charge Generation Layer*, IDMC 2003, Taiwan; Session 21 OLED (5)). The materials in these layers may also be doped. Each of these layers does not necessarily have to be present. Suitable hole-transport materials are, for example, aromatic amines, as usually used in accordance with the prior art and which may also be p-doped. Suitable electron-transport materials are, for example, metal chelate complexes, for example AlQ_3, compounds based on electron-deficient heterocycles, for example triazine derivatives, or compounds containing aromatic carbonyls or phosphine oxides, as described, for example, in WO 05/084081 and WO 05/084082, which may in each case also be n-doped. Suitable electron-injection materials are, in particular, fluorides and oxides of the alkali and alkaline earth metals, for example NaF, BaF_2, CaF_2, LiF or Li_2O.

In a preferred embodiment of the invention, the compound of the formula (1) is employed as host material, in particular for fluorescent emitters, and/or as electron-transport material and/or as hole-blocking material. This is the case, in particular, if the compound does not contain any substituents of the formula $N(Ar^1)_2$.

A host material is taken to mean the component in a system comprising host and dopant (binary mixture) which is present in the system in the higher proportion. In a system comprising a host and a plurality of dopants (ternary and higher mixtures), the host is taken to mean the component whose proportion is the highest in the mixture.

The proportion of the host material of the formula (1) in the emitting layer is between 50.0 and 99.9% by weight, preferably between 80.0 and 99.5% by weight, particularly preferably between 90.0 and 99.0% by weight. Correspondingly, the proportion of the dopant in the emitting layer is between 0.1 and 50.0% by weight, preferably between 0.5 and 20.0% by weight, particularly preferably between 1.0 and 10.0% by weight.

Preferred dopants are selected from the class of the aromatic anthracenamines, the aromatic anthracenediamines, the aromatic pyrenamines, the aromatic pyrenediamines, the monostyrylamines, the distyrylamines, the tristyrylamines, the tetrastyrylamines, the styrylphosphines, the styryl ethers and the arylamines. An aromatic anthracenamine is taken to mean a compound in which a diarylamino group is bonded directly to an anthracene group, preferably in the 9-position. An aromatic anthracenediamine is taken to mean a compound in which two diarylamino groups are bonded directly to an anthracene group, preferably in the 9,10-position. Aromatic pyrenamines and pyrenediamines are defined analogously, with the diarylamino groups preferably being bonded to the pyrene in the 1-position or in the 1,6-position. A monostyrylamine is taken to mean a compound which contains a substituted or unsubstituted styryl group and at least one, preferably aromatic, amine. A distyrylamine is taken to mean a compound which contains two substituted or unsubstituted styryl groups and at least one, preferably aromatic, amine. A tristyrylamine is taken to mean a compound which contains three substituted or unsubstituted styryl groups and at least one, preferably aromatic, amine. A tetrastyrylamine is taken to mean a compound which contains four substituted or unsubstituted styryl groups and at least one, preferably aromatic, amine. Corresponding phosphines and ethers are defined analogously to the amines. For the purposes of this invention, an arylamine or an aromatic amine is taken to mean a compound which contains three substituted or unsubstituted aromatic or heteroaromatic ring systems bonded directly to the nitrogen. At least one aryl group here is preferably a condensed aryl group having at least three rings. The styryl groups are particularly preferably stilbenes, which may also be further substituted. Particularly preferred dopants are selected from the classes of the tristilbenamines, the aromatic stilbenediamines, the anthracenediamines and the pyrenediamines, Very particularly preferred dopants are selected from the class of the tristyrylamines. Examples of dopants of this type are substituted or unsubstituted tristilbenamines or the dopants described in WO 06/000388, WO 06/058737 and WO 06/000389.

In a further embodiment of the invention, the organic electroluminescent device comprises a plurality of emitting layers, where at least one of these layers comprises at least one compound of the formula (1). These emission layers particularly preferably have in total a plurality of emission maxima between 380 nm and 750 nm, resulting overall in white emission, i.e. at least one further emitting compound which is able to fluoresce or phosphoresce and emits yellow, orange or red light is used in the further emitting layer(s). Preference is given to three-layer systems, where at least one of these layers comprises at least one compound of the formula (1) and where the three layers exhibit blue, green and orange or red emission (for the basic structure, see, for example, WO 05/011013). Emitters which have broad-band emission bands and thus exhibit white emission are likewise suitable for white emission.

In addition to the compounds of the formula (1) and the dopant(s), further substances, for example hole- or electron-transport materials, may also be present in the emitting layer.

If the symbol R stands for an $N(Ar^1)_2$ group and/or at least one substituent R^1 on the Ar group or in another position stands for an $N(Ar^1)_2$ group, the compound of the formula (1) is particularly suitable as emitting compound and/or as hole-transport material, as described in more detail below.

If the compound of the formula (1) is employed as hole-transport material, it is preferably employed in a hole-transport layer and/or in a hole-injection layer. For the purposes of this invention, a hole-injection layer is a layer which is directly adjacent to the anode. For the purposes of this invention, a hole-transport layer is a layer which is located between a hole-injection layer and an emission layer. If the compounds of the formula (1) are used as hole-transport or hole-injection material, it may be preferred for them to be doped with electron-acceptor compounds, for example with F_4-TCNQ or with compounds as described in EP 1476881 or EP 1596445.

If the compound of the formula (1) is employed as emitting compound, it is preferably employed in combination with a host material.

The proportion of the emitting compound of the formula (1) in the mixture of the emitting layer is between 0.1 and 50.0% by weight, preferably between 0.5 and 20.0% by weight, particularly preferably between 1.0 and 10.0% by weight. Correspondingly, the proportion of the host material in the layer is between 50.0 and 99.9% by weight, preferably between 80.0 and 99.5% by weight, particularly preferably between 90.0 and 99.0% by weight.

Suitable host materials are various classes of substance. Preferred host materials are selected from the classes of the oligoarylenes (for example 2, 2',7,7'-tetraphenylspirobifluorene as described in EP 676461 or dinaphthylanthracene), in particular the oligoarylenes containing condensed aromatic groups, the oligoarylenevinylenes (for example DPVBi or spiro-DPVBi as described in EP 676461), the polypodal metal complexes (for example as described in WO 04/081017), the hole-conducting compounds (for example as described in WO 04/058911), the electron-conducting compounds, in particular ketones, phosphine oxides, sulfoxides, etc. (for example as described in WO 05/084081 or WO 05/084082), the atropisomers (for example as described in WO 06/048268) or the boronic acid derivatives (for example as described in WO 06/117052). Particularly preferred host materials are selected from the classes of the oligoarylenes containing naphthalene, anthracene and/or pyrene or atropisomers of these compounds, the oligoarylenevinylenes, the ketones, the phosphine oxides and the sulfoxides. Very particularly preferred host materials are selected from the classes of the oligoarylenes containing anthracene and/or pyrene or atropisomers of these compounds, the phosphine oxides and the sulfoxides.

Preference is furthermore given to an organic electroluminescent device, characterised in that one or more layers are coated by means of a sublimation process, in which the materials are vapour-deposited in vacuum sublimation units at a pressure of less than 10^{-5} mbar, preferably less than 10^{-6} mbar, particularly preferably less than 10^{-7} mbar.

Preference is likewise given to an organic electroluminescent device, characterised in that one or more layers are coated by the OVPD (organic vapour phase deposition) process or with the aid of carrier-gas sublimation, in which the materials are applied at a pressure between 10^{-5} mbar and 1 bar.

Preference is furthermore given to an organic electroluminescent device, characterised in that one or more layers are produced from solution, such as, for example, by spin coating, or by means of any desired printing process, such as, for example, screen printing, flexographic printing or offset printing, but particularly preferably LITI (light-induced thermal imaging, thermal transfer printing) or ink-jet printing. Soluble compounds of the formula (1) are necessary for this purpose. The compounds according to the invention are therefore very highly suitable for processing from solution since, due to the substitution, they have high solubility in organic solvents.

The organic electroluminescent devices according to the invention have the following surprising properties:

1. The compounds according to the invention have high thermal stability and in particular a high glass-transition temperature.
2. The compounds according to the invention exhibit high efficiency, a good lifetime and good colour coordinates on use in OLEDs.
3. The compounds according to the invention have good solubility in organic solvents, which simplifies the preparation and processing of these compounds.
4. The compounds according to the invention have high redox stability (high stability to holes and electrons).
5. The film-formation properties of the compounds according to the invention are very good.

The present application text is directed to the use of compounds according to the invention in relation to OLEDs and the corresponding displays.

In spite of this restriction of the description, it is possible for the person skilled in the art, without further inventive step, also to use the compounds according to the invention for further uses in other electronic devices, for example for organic field-effect transistors (O-FETs), organic thin-film transistors (O-TFTs), organic light-emitting transistors (O-LETs), organic integrated circuits (O-ICs), organic solar cells (O-SCs), organic field-quench devices (O-FQDs), organic photo receptors, light-emitting electrochemical cells (LECs) or also organic laser diodes (O-lasers), to mention but a few applications.

The present invention furthermore relates to the use of the compounds according to the invention in the corresponding devices and to these devices themselves.

The invention is explained in greater detail by the following examples, without wishing to restrict it thereby.

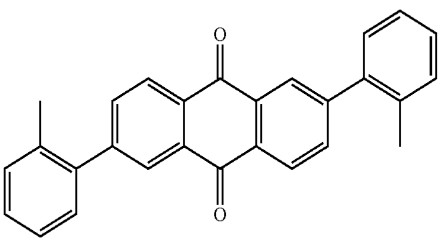
EXAMPLES

The following syntheses are carried out under a protective-gas atmosphere, unless indicated otherwise. The starting materials can be purchased from ALDRICH or ABCR (palladium(II) acetate, tri-o-tolyl-phosphine, di-tert-butylchlorophosphine, bromides, amines, inorganics, solvents). 2,6-Dibromoanthraquinone is prepared by the method of Lee et al., *Org. Lett.* 2005, 7(2), 323; 2-trimethylsilylbromobenzene is prepared by the method of Klusener et al., *Org. Chem.* 1990, 55(4), 1311; pinacolyl 10-(4-methylnaphth-1-yl)anthracene-9-boronate is prepared in accordance with EP 05009643.7; 1-bromo-2-(1-methyl-1-phenylethyl)benzene is prepared by the method of Sigmundova et al., *Synth. Commun.* 2004, 34(20), 3667.

Example 1

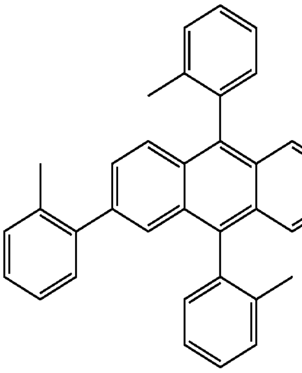
2,6,9,10-Tetra-o-tolylanthracene

a) 2,6-Bis-o-tolylanthraquinone



A suspension of 28.7 g (100 mmol) of 2,6-dibromoanthraquinone, 32.6 g (240 mmol) of o-tolylboronic acid, 89.2 g (420 mmol) of potassium phosphate, 1.8 g (8 mmol) of tri-o-tolylphosphine and 225 mg (1 mmol) of palladium(II) acetate in a mixture of 200 ml of dioxane, 400 ml of toluene and 500 ml of water is refluxed for 16 h. After cooling, the solid is filtered off with suction, washed three times with 100 ml of water each time and three times with 100 ml of ethanol each time, dried in vacuo and subsequently recrystallised twice from DMF. Yield: 33.0 g (85 mmol), 84.9% of theory, purity: 98% according to NMR.

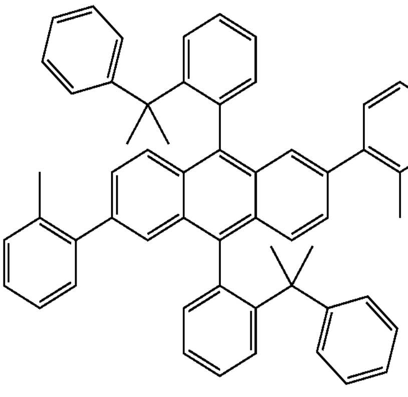
b) 2,6,9,1-Tetra-o-tolylanthracene



The corresponding Grignard reagent is prepared from 3.7 g (153 mmol) of magnesium and 18.0 ml (150 mmol) of 2-bromotoluene in 500 ml of THF. 19.4 g (50 mmol) of 2,6-bis-o-tolylanthraquinone are added to the Grignard reagent. The reaction mixture is subsequently refluxed for 16 h. After cooling, 30 ml of ethanol are added, the solvent is removed in vacuo, the residue is taken up in 300 ml of DMF and warmed to 60° C., and 8.9 g (65 mmol) of tin(II) chloride are then added in portions with vigorous stirring (note: exothermic reaction!). The mixture is subsequently stirred at 60° C. for a further 2 h. After cooling, 500 ml of 2.5N hydrochloric acid are added, and the solid is filtered off with suction. The solid is washed three times with 100 ml of 2.5N hydrochloric acid each time, three times with 100 ml of water each time and three times with 100 ml of ethanol each time, dried in vacuo and subsequently recrystallised three times from acetic acid and twice from DMF. Sublimation $p=1\times10^{-5}$ mbar, T=335° C. Yield: 19.8 g (37 mmol), 73.5% of theory; purity: 99.8% according to HPLC. Mixture of two atropisomers according to ^{1}H-NMR spectroscopy.

Example 2

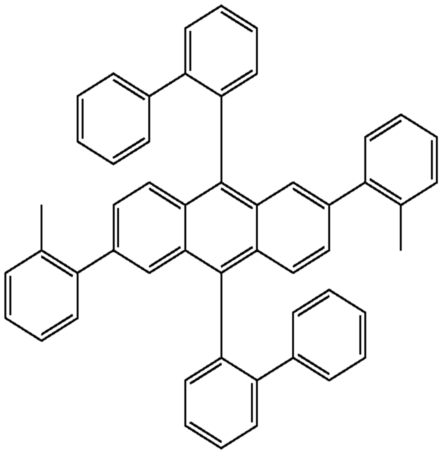
2,6-Bis-o-tolyl-9,10-bis(2-(1-methyl-1-phenylethyl)phenyl)-anthracene



Procedure analogous to Example 1b. Instead of 18.0 ml (150 mmol) of 2-bromotoluene, 41.3 g (150 mmol) of 1-bromo-2-(1-methyl-1-phenylethyl)benzene are used. Recrystallisation from dioxane. Sublimation $p=1\times10^{-5}$ mbar, T=360° C. Yield: 22.6 g (30 mmol), 60.5% of theory; purity: 99.9% according to HPLC. Atropisomerically pure according to ^{1}H-NMR spectroscopy.

Example 3

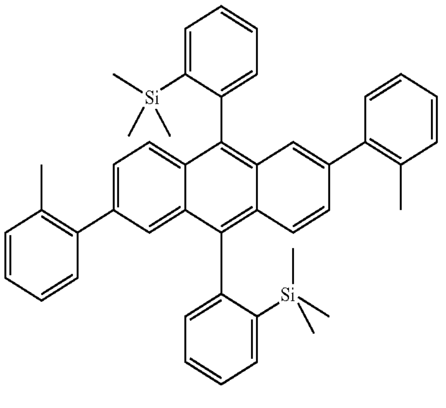
2,6-Bis-o-tolyl-9,10-bis(2-biphenyl)anthracene



Procedure analogous to Example 1b. Instead of 18.0 ml (150 mmol) of 2-bromotoluene, 25.9 ml (150 mmol) of 2-bromobiphenyl are used. Recrystallisation from chlorobenzene. Sublimation $p=1\times10^{-5}$ mbar, T=360° C. Yield: 27.1 g (41 mmol), 81.7% of theory; purity: 99.9% according to HPLC. Atropisomerically pure according to ^{1}H-NMR spectroscopy.

Example 4

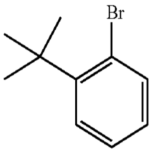
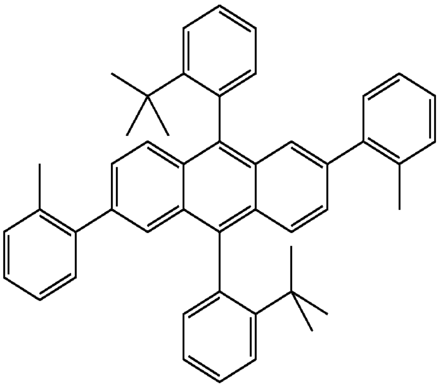
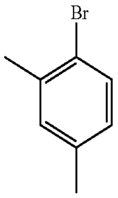
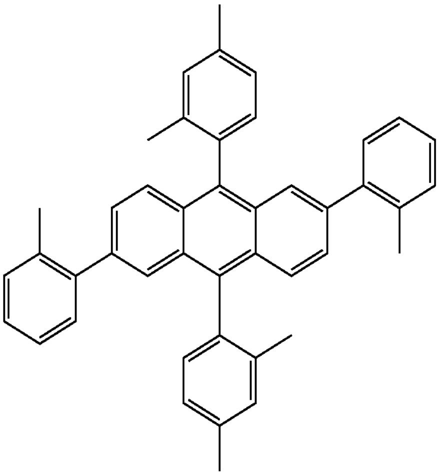
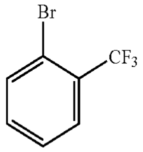
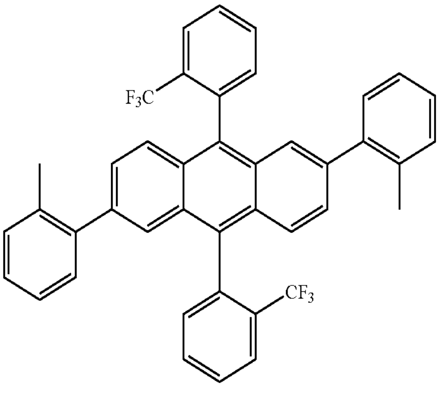
2,6-Bis-o-tolyl-9,10-bis(2-trimethylsilylphenyl)anthracene



Procedure analogous to Example 1b. Instead of 18.0 ml (150 mmol) of 2-bromotoluene, 34.4 g (150 mmol) of 2-trimethylsiylbromobenzene are used. Recrystallisation from dioxane. Sublimation p=1×10^{-5} mbar, T=330° C. Yield: 21.9 g (33 mmol), 66.8% of theory; purity: 99.9% according to HPLC. Atropisomerically pure according to ^{1}H-NMR spectroscopy.

Example 5

The following compounds are prepared analogously to Examples 1b, 2, 3 and 4:

Ex.	Bromide	Product
6		
7		
8		

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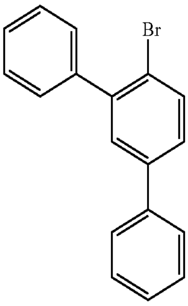
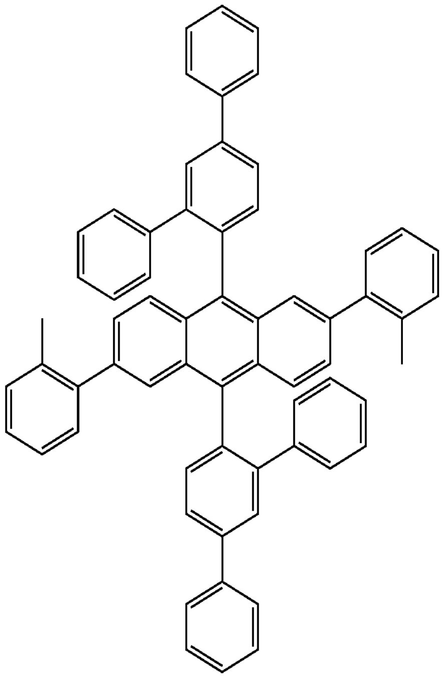
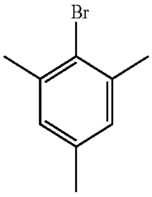
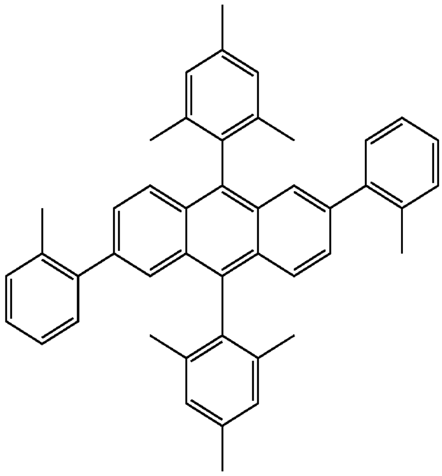
Ex.	Bromide	Product
9		
10		

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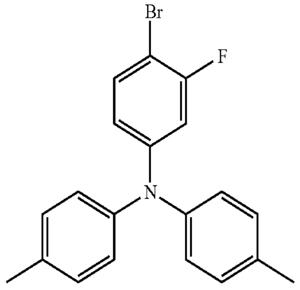
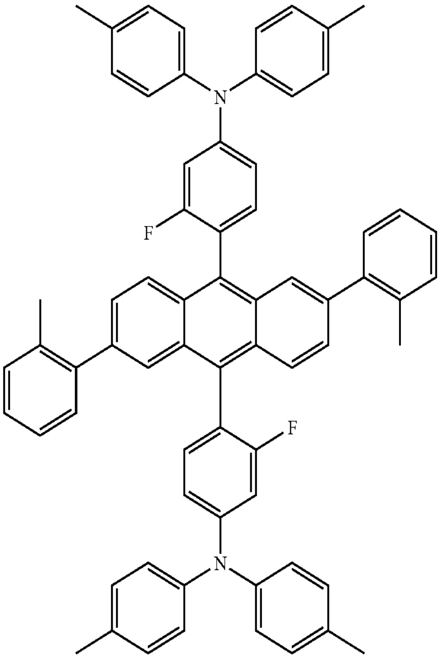
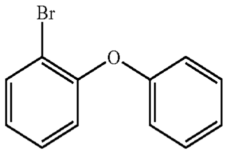
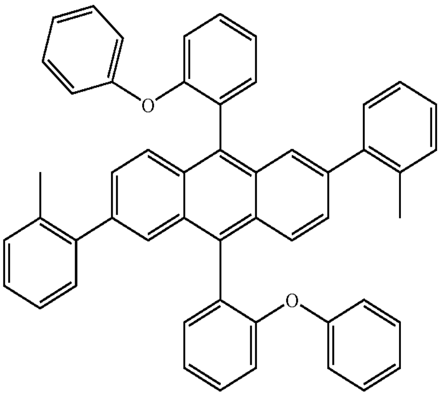
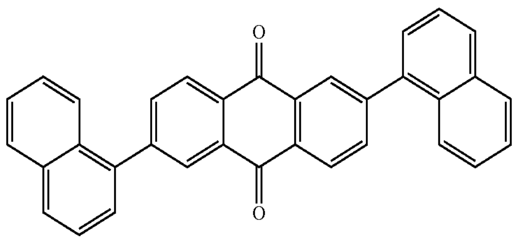
Ex.	Bromide	Product
11		
12		

Example 13

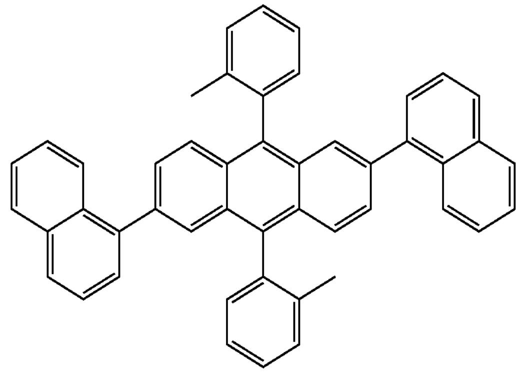
2,6-Bisnaphth-1-yl-9,10-bis-o-tolylanthracene

a) 2,6-Bisnaphth-1-ylanthraquinone



A suspension of 28.7 g (100 mmol) of 2,6-dibromoanthraquinone, 44.7 g (260 mmol) of 1-naphthylboronic acid, 89.2 g (420 mmol) of potassium phosphate, 1.8 g (6 mmol) of tri-o-tolylphosphine and 225 mg (1 mmol) of palladium(II) acetate in a mixture of 200 ml of dioxane, 400 ml of toluene and 500 ml of water is refluxed for 16 h. After cooling, the solid is filtered off with suction, washed three times with 100 ml of water each time and three times with 100 ml of ethanol each time, dried in vacuo and subsequently recrystallised twice from chlorobenzene. Yield: 41.6 g (90 mmol), 90.3% of theory; purity: 99% according to NMR.

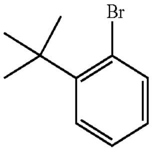
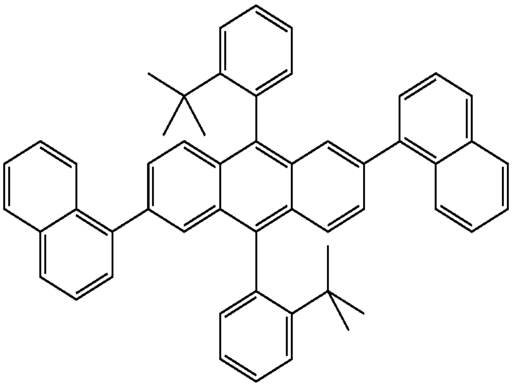
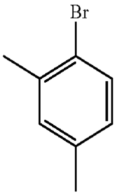
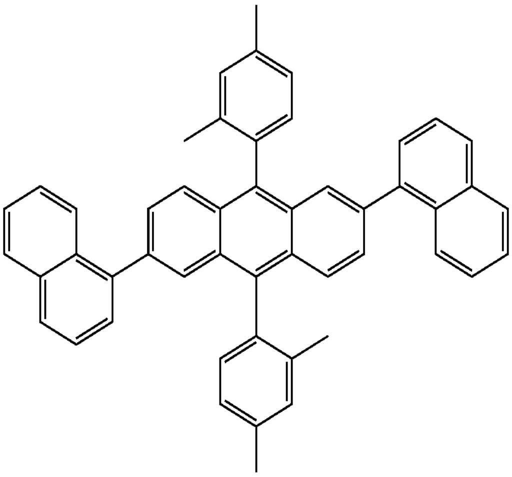
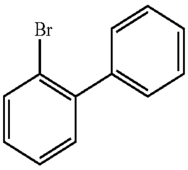
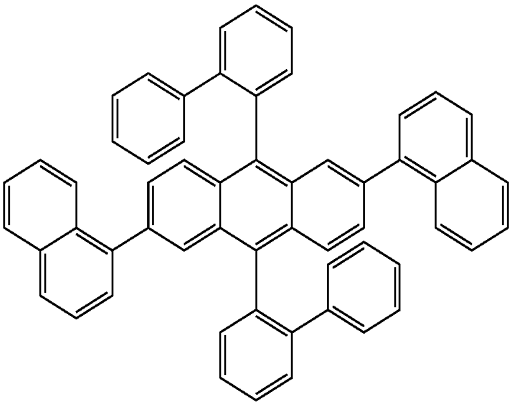
b) 2,6-Bisnaphth-1-yl-9,10-bis-o-tolylanthracene



Procedure analogous to Example 1b. Recrystallisation from NMP. Sublimation $p=1\times10^{-5}$ mbar, T=375° C. Yield: 22.2 g (36 mmol), 72.7% of theory; purity: 99.9% according to HPLC. Mixture of two atropisomers according to ^{1}H-NMR spectroscopy.

Example 14

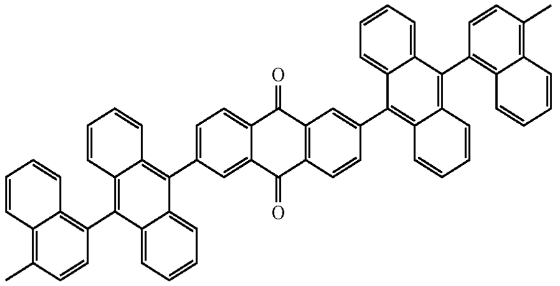
The following compounds are prepared analogously to Example 13:

Ex.	Bromide	Product
15		
16		
17		

Example 18

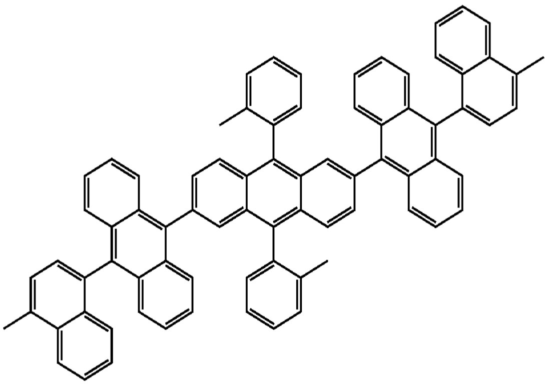
2,6-Bis(9-(4-methylnaphthyl)anthracen-10-yl)-9,10-bis-o-tolylanthracene

a) 2,6-Bis-(9-(4-methylnaphthyl)anthracen-10-yl) anthraquinone



A suspension of 28.7 g (100 mmol) of 2,6-dibromoanthraquinone, 133.3 g (300 mmol) of pinacolyl 10-(4-methylnaphth-1-yl)anthracene-9-boronate, 96.7 g (600 mmol) of potassium fluoride and 1.2 g (1 mmol) of tetrakis-triphenylphosphinopalladium(0) in a mixture of 500 ml of ethylene glycol dimethyl ether, 200 ml of ethanol and 400 ml of water is refluxed for 36 h. After cooling, the solid is filtered off with suction, washed three times with 100 ml of water each time and three times with 100 ml of ethanol each time, dried in vacuo and subsequently recrystallised twice from o-dichlorobenzene. Yield: 66.9 g (79 mmol), 79.5% of theory; purity: 98% according to NMR.

b) 2,6-Bis-(9-(4-methylnaphthyl)anthracen-10-yl)-9,10-bis-o-tolyl-anthracene

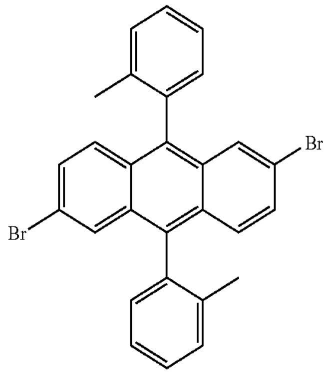


Procedure analogous to Example 1b. Instead of 19.4 g (50 mmol) of 2,6-bis-o-tolylanthraquinone, 42.1 g (50 mmol) of 2,6-bis(9-(4-methylnaphthyl) anthracen-10-yl)anthraquinone are used. After addition of the 2,6-bis(9-(4-methylnaphthyl)anthracen-10-yl)anthraquinone, 300 ml of toluene are added to the reaction mixture. Recrystallisation from o-dichlorobenzene. Sublimation $p=1\times10^{-5}$ mbar, T 400° C. Yield: 27.5 g (28 mmol), 55.5% of theory; purity: 99.9% according to HPLC. Mixture of two atropisomers according to ^{1}H-NMR spectroscopy.

Example 19

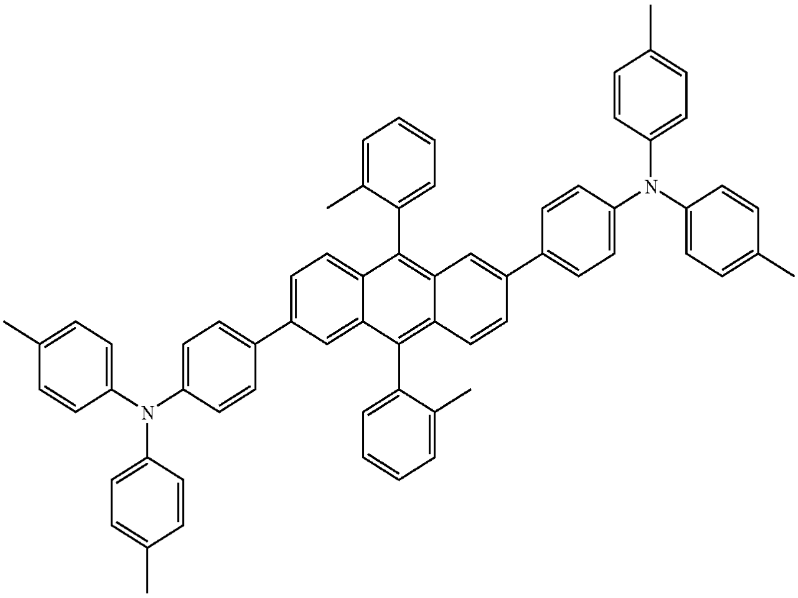
2,6-Bis(p-tolylamino)-9,10-bis-o-tolylanthracene

a) 2,6-Dibromo-9,10-bis-o-tolylanthracene



Procedure analogous to Example 1b. Instead of 19.4 g (50 mmol) of 2,6-bis-o-tolylanthraquinone, 18.3 g (50 mmol) of 2,6-dibromoanthraquinone are used. Recrystallisation from toluene. Yield: 12.3 g (24 mmol), 47.6% of theory; purity: 97% according to NMR.

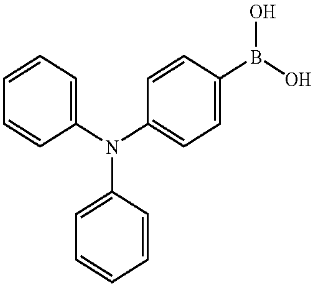
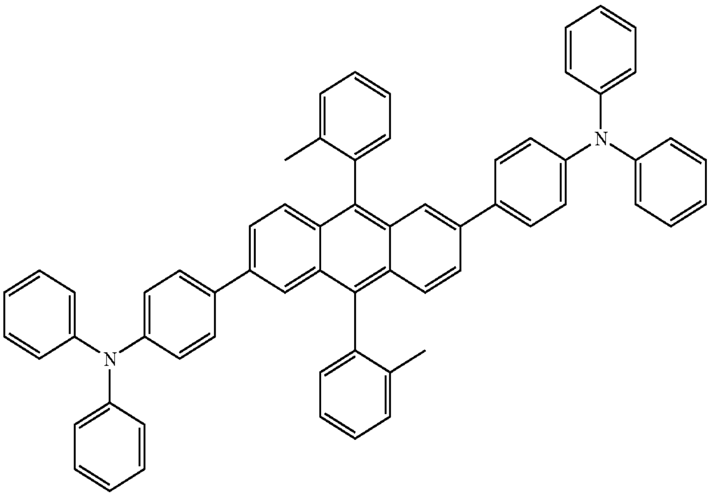
b) 2,6-Bis(di-p-tolylaminophenyl-4-yl)-9,10-bis-o-tolylanthracene



A suspension of 51.6 g (100 mmol) of 2,6-dibromo-9,10-bis-o-tolylanthracene, 82.5 g (260 mmol) of di-p-tolylaminophenyl-4-boronic acid, 89.2 g (420 mmol) of potassium phosphate, 1.8 g (6 mmol) of tri-o-tolylphosphine and 225 mg (1 mmol) of palladium(II) acetate in a mixture of 200 ml of dioxane, 400 ml of toluene and 500 ml of water is refluxed for 16 h. After cooling, the solid is filtered off with suction, washed three times with 100 ml of water each time, washed three times with 100 ml of ethanol each time, dried in vacuo, recrystallised five times from DMF and then sublimed in vacuo ($p=1\times10^{-5}$ mbar, T=365° C.). Yield: 68.7 g (76 mmol), 76.2% of theory; purity: 99.9% according to HPLC.

Example 20

The following compounds are prepared analogously to Example 19:

Ex.	Amine	Product
21		

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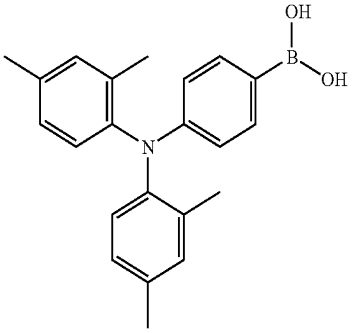
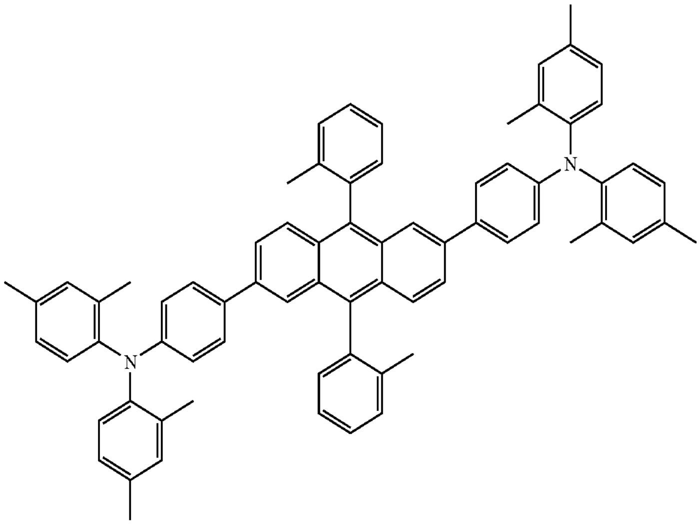
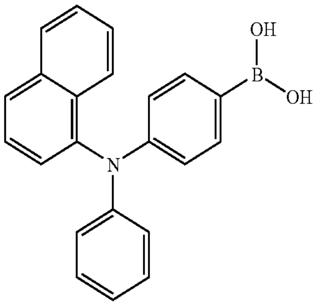
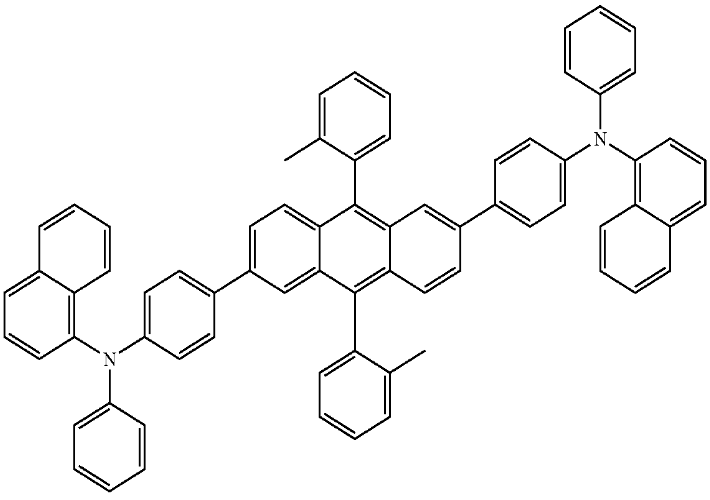
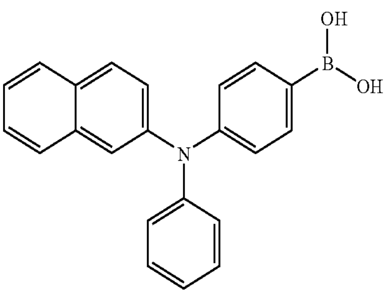
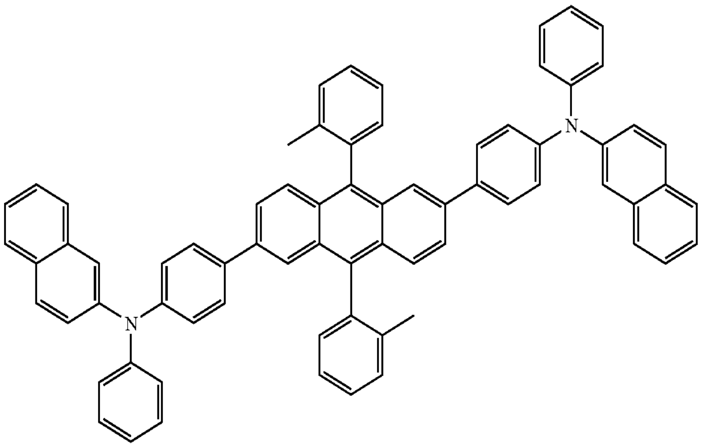
Ex.	Amine	Product
22		
23		
24		

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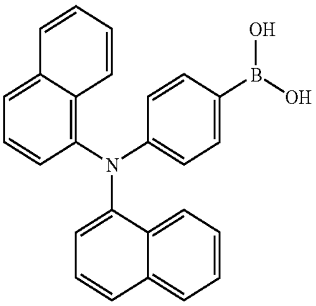
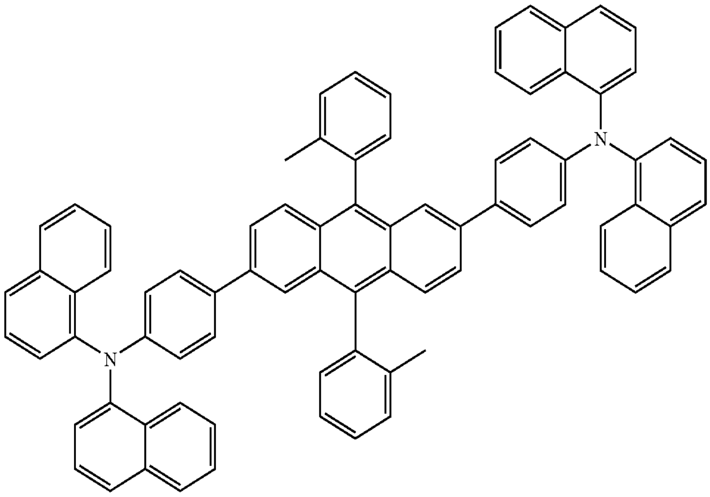
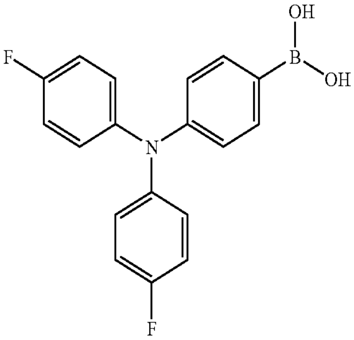
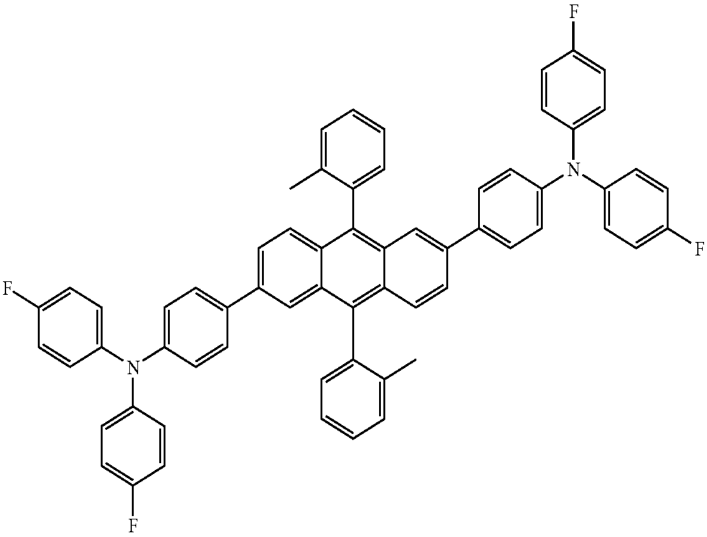
Ex.	Amine	Product
25		
26		

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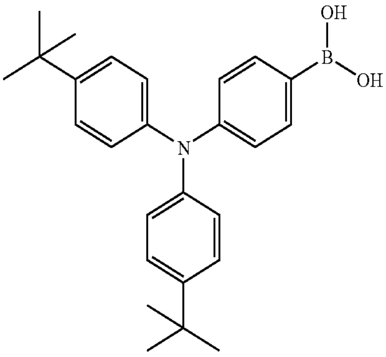
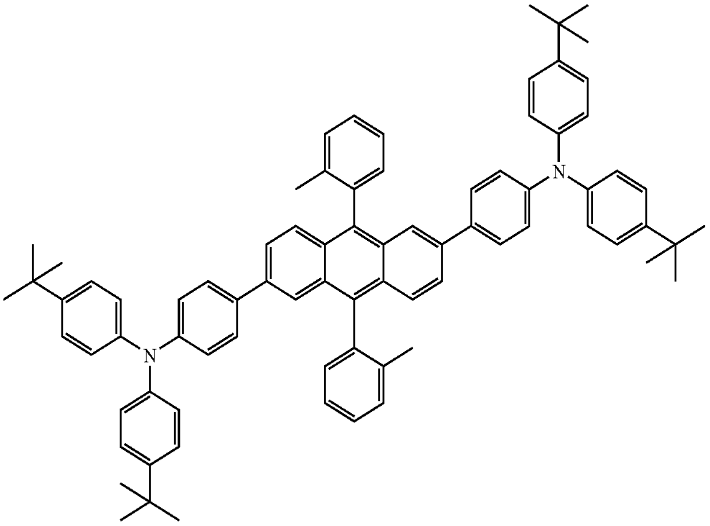
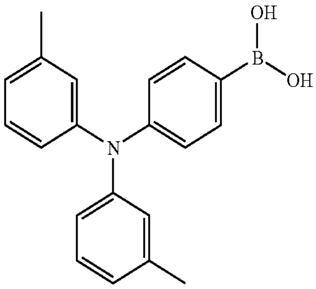
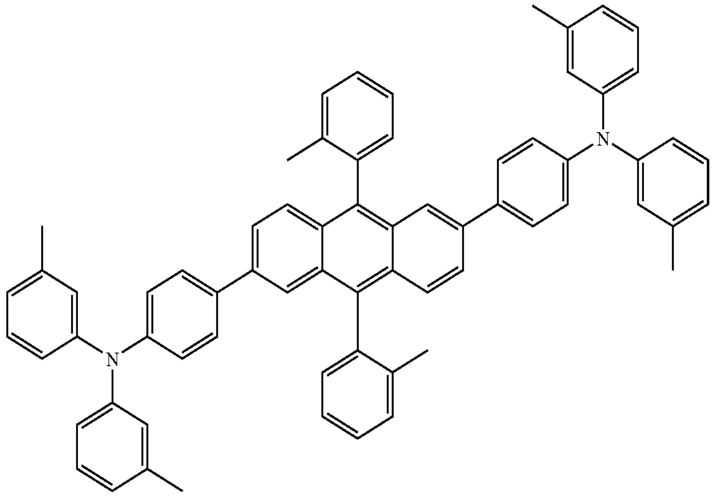
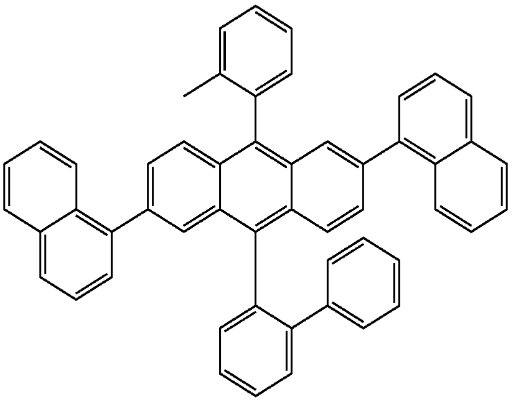
Ex.	Amine	Product
27		
28		

Example 29

2,6-Bisnaphth-1-yl-9-o-tolyl-10-2-biphenylanthracene

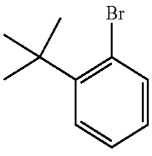
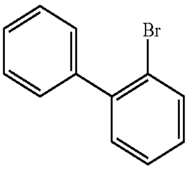
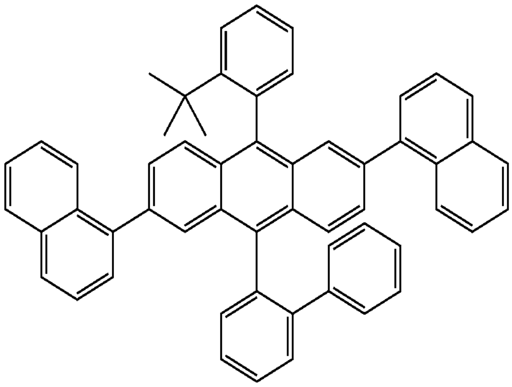
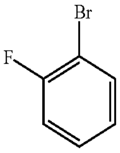
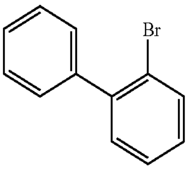
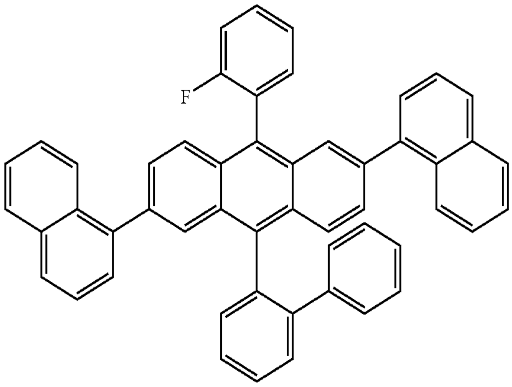
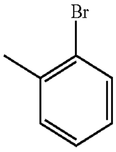
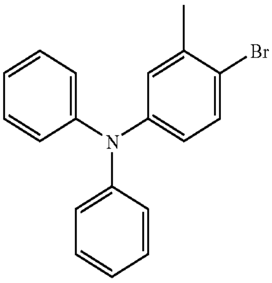
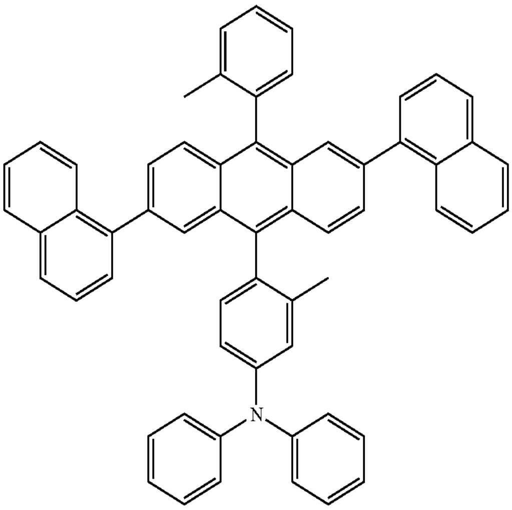


A 2-lithiobiphenyl solution in 200 ml of THF, prepared from 11.7 g (50 mmol) of 2-bromobiphenyl and 20 ml (50 mmol) of 2.5N n-butyllithium at −78° C., is added dropwise at −78° C. with vigorous stirring to a suspension of 46.1 g (100 mmol) of 2,6-bisnaphth-1-ylanthraquinone in 500 ml of THF, and the mixture is stirred for a further 30 min. A 2-lithiotoluene solution in THF, prepared from 8.7 g (50 mmol) of 2-bromotoluene and 20 ml (50 mmol) of 2.5 N n-butyllithium at −78° C., is subsequently added to this suspension, and the mixture is stirred for a further 30 min. The reaction mixture is allowed to warm to room temperature, 30 ml of ethanol are added, and the solvent is removed in vacuo. The residue is taken up in 300 ml of DMF and warmed to 60° C., and 17.7 g (130 mmol) of tin(II) chloride are added in portions with vigorous stirring (note: exothermic reaction!). The mixture is subsequently stirred at 60° C. for a further 2 h. After cooling, 500 ml of 2.5 N hydrochloric acid are added, and the solid is filtered off with suction. The solid is washed three times with 100 ml of 2.5 N hydrochloric acid each time, three times with 100 ml of water each time and three times with 100 ml of ethanol each time, dried in vacuo and subsequently recrystallised once from acetic acid and three times from dioxane.

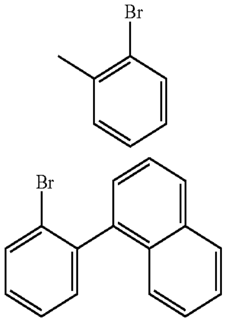
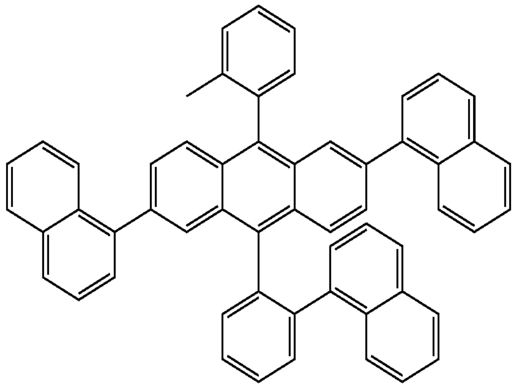
Sublimation p=1×10^{-5} mbar, T=345° C. Yield: 43.1 g (64 mmol), 64.0% of theory; purity: 99.9% according to HPLC. Mixture of two atropisomers according to ^{1}H-NMR spectroscopy.

Example 30

The following compounds are prepared analogously to Example 29:

Ex.	Bromides	Product
31	 	
32	 	
33	 	

-continued

Ex.	Bromides	Product
34	 	

Example 35

Production of OLEDs

OLEDs are produced by a general process as described in WO 04/058911, which is adapted in individual cases to the particular circumstances (for example layer-thickness variation in order to achieve optimum efficiency or colour).

The results for various OLEDs are presented in Examples 36 to 48 below. Glass plates coated with structured ITO (indium tin oxide) form the substrates of the OLEDs. For improved processing, PEDOT (spin-coated from water; purchased from H. C. Starck, Goslar, Germany; poly(3,4-ethylene-dioxy-2,5-thiophene)) is applied directly to the substrate. The OLEDs always consist of the following layer sequence: substrate/PEDOT 20 nm/hole-injection layer (HIL1) 20 nm/hole-transport layer (HTM1) 20 nm/emission layer (EML) 30 nm/electron-transport layer (ETM1) 20 nm and finally a cathode. The materials apart from PEDOT are thermally vapour-deposited in a vacuum chamber. The EML here always consists of a matrix material (host) and a dopant (guest), which is admixed with the host by co-evaporation. The cathode is formed by a 1 nm thin LiF layer and a 150 nm Al layer deposited on top. Table 2 shows the chemical structures of the materials used to construct the OLEDs.

These OLEDs are characterised by standard methods; for this purpose, the electroluminescence spectra, the efficiency (measured in cd/A), the power efficiency (measured in Im/W) as a function of the brightness, calculated from current/voltage/luminance characteristics (IUL characteristics), and the lifetime are determined

+. The lifetime is defined as the time after which the initial luminance has dropped from 1000 cd/m^2 to half.

Table 3 shows the results for some OLEDs (Examples 36 to 48). The comparative example used is host H1 in accordance with the prior art.

TABLE 2

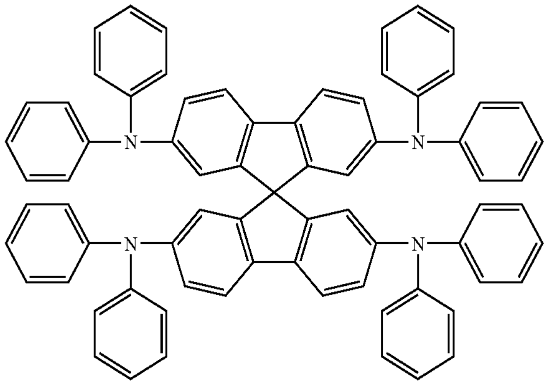
Compounds used
   HIL1

TABLE 2-continued

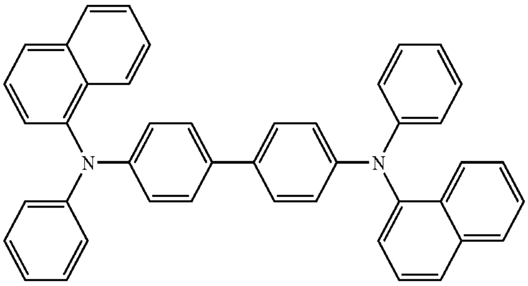
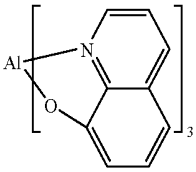
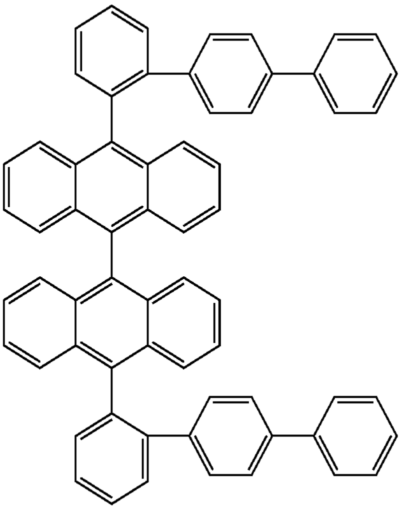
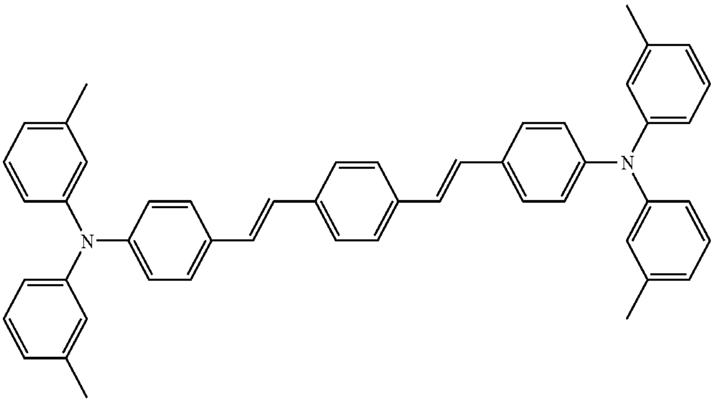
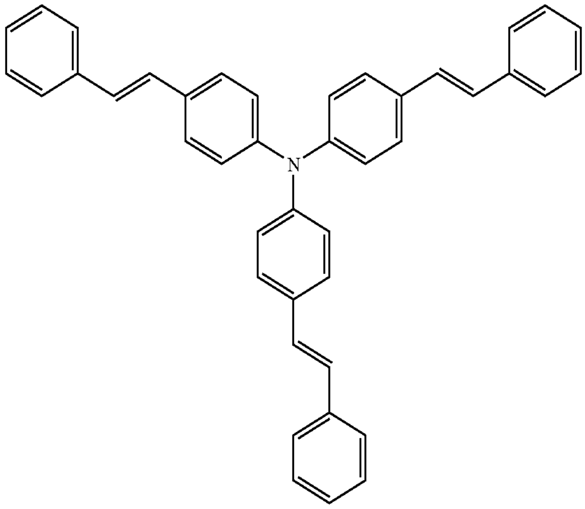
Compounds used
 HTM1
 ETM1
 H1 (comparison)
 D1

TABLE 2-continued

Compounds used



D2

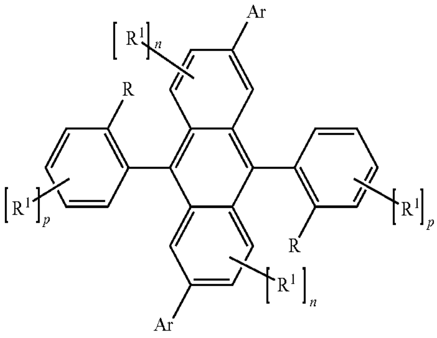
TABLE 3

OLED results					
Example	EML	Max. efficiency (cd/A)	Voltage (V) at 1000 cd/m^2	CIE	Lifetime (h) at 1000 cd/m^2
36 (comparison)	H1 + 5% D1	9.9	5.7	x = 0.17/y = 0.33	4050
37 (comparison)	H1 + 5% D2	3.4	6.2	x = 0.15/y = 0.13	1200
38	Ex. 1 + 5% D1	10.5	5.5	x = 0.17/y = 0.33	6100
39	Ex. 1 + 5% D2	3.8	5.8	x = 0.15/y = 0.14	1800
40	Ex. 3 + 5% D1	12.2	5.7	x = 0.17/y = 0.33	5800
41	Ex. 3 + 5% D2	4.2	5.9	x = 0.15/y = 0.14	1600
42	Ex. 9 + 5% D2	11.3	5.4	x = 0.17/y = 0.32	6300
43	Ex. 9 + 5% D2	3.9	5.8	x = 0.15/y = 0.15	2200
44	Ex. 17 + 5% D3	11.5	5.5	x = 0.17/y = 0.33	7100
45	Ex. 17 + 5% D3	3.5	5.9	x = 0.15/y = 0.14	2000
46	Ex. 9 + 5% Ex. 11	7.8	5.3	x = 0.15/y = 0.19	4800
47	Ex. 9 + 5% Ex. 19	8.0	5.6	x = 0.15/y = 0.24	5300
48	Ex. 17 + 7% Ex. 19	8.3	5.3	x = 0.16/y = 0.26	5600

The invention claimed is:

1. A compound of formula (1)

Formula (1)



wherein

Ar is, identically or differently on each occurrence, phenyl, 1-naphthyl, 2-naphthyl, 2-anthryl, 9-anthryl, 2-phenanthrenyl, 3-phenanthrenyl or 9-phenanthrenyl each optionally substituted by one or more radicals R';

R is, identically on each occurrence, F, Si(R^2)$_3$; —N(Ar^1)$_2$; a straight-chain alkyl or alkoxy group having 1 to 4 C atoms or a branched alkyl group having 3 to 5 C atoms, each of which may be substituted by one or more radicals R^2, where one or more H atoms may be replaced by F, or a phenyl group;

R^1

are, identically or differently on each occurrence, F; Cl; Br; I; CN; N(Ar^1)$_2$; C(═O)Ar^1; P(Ar^1)$_2$; P(═O)(Ar^1)$_2$; Si(R^2)$_3$; NO$_2$; a straight-chain alkyl, alkoxy or thioalkoxy group having 1 to 40 C atoms optionally substituted by one or more radicals R^2; or a branched or cyclic alkyl, alkoxy or thioalkoxy group having 3 to 40 C atoms optionally substituted by one or more radicals R^2; wherein one or more non-adjacent CH$_2$ groups of said straight-chain alkyl, alkoxy or thioalkoxy group or said branched or cyclic alkyl, alkoxy or thioalkoxy group are optionally replaced by —R^{2}C═CR^2—, —C≡C—, Si(R^2)$_2$, Ge(R^2)$_2$, Sn(R^2)$_2$, C═O, C═S, C═Se, C═NR^2, —O—,

—S—, —N(R^2)—, or —CONR^2— and wherein one or more H atoms of said straight-chain alkyl, alkoxy or thioalkoxy group or said branched or cyclic alkyl, alkoxy or thioalkoxy group are optionally replaced by F, Cl, Br, I, CN, or NO_2; an aromatic or hetero-aromatic ring system having 5 to 30 aromatic ring atoms optionally substituted by one or more radicals R^2; or an aryloxy or heteroaryloxy group having 5 to 24 aromatic ring atoms optionally substituted by one or more radicals R^2; or a combination of two, three, four or five substituents R^1;

Ar^1 is, identically or differently on each occurrence, an aromatic or heteroaromatic ring system having 5 to 30 aromatic ring atoms optionally substituted by one or more non-aromatic radicals R^1 and wherein two radicals Ar^1 are optionally connected to one another by a single bond or an O, S, N(R^2), or C(R^2)$_2$ group;

R^2 is, identically or differently on each occurrence, H or a hydrocarbon radical having 1 to 20 C atoms, wherein said hydrocarbon radical is aliphatic, aromatic, or a combination of aliphatic and aromatic and is optionally substituted by F and wherein two or more radicals R^2 optionally define a mono- or polycyclic, aliphatic or aromatic ring system with one another;

n is 0;

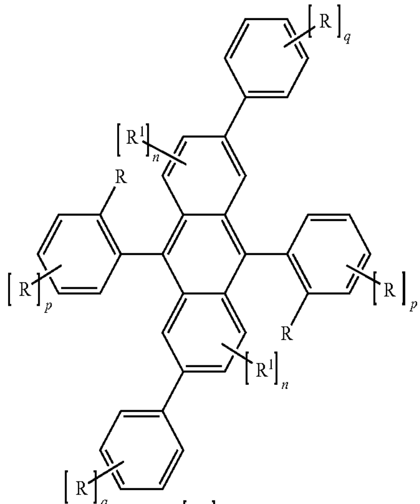
p is 0;

with the proviso that if R^1 contains a benzimidazole group, it is not bonded to Ar.

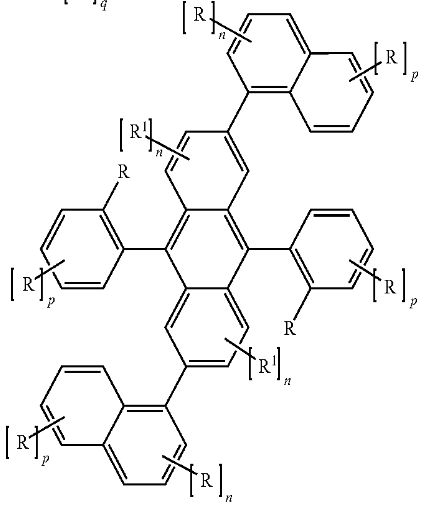
2. The compound of claim **1**, wherein both Ar are identical.

3. The compound of claim **1**, wherein said compound has a formula selected from the group consisting of formulae (2), (3), (4), and (5)

Formula (2)



Formula (3)



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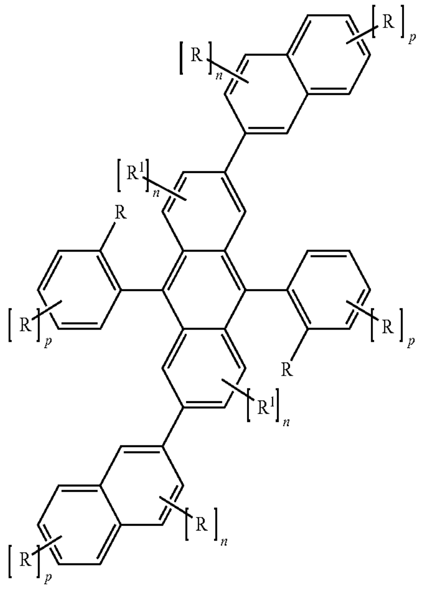
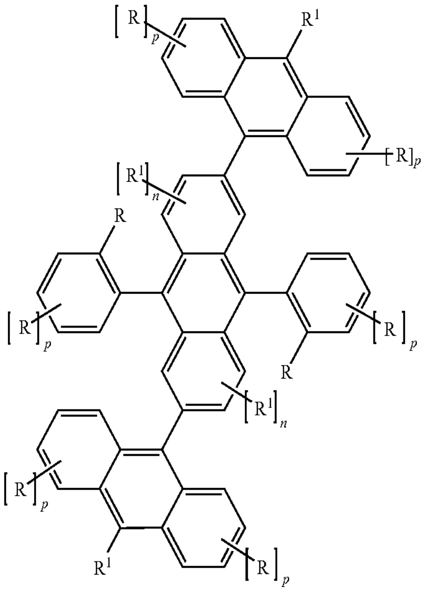
Formula (4)



Formula (5)



wherein q is 0, 1, 2, 3, 4, or 5.

4. A process for preparing the compound of claim **1** comprising (1) reacting an anthraquinone substituted in the 2,6-position by chlorine, bromine, iodine, or a sulfonic acid derivative with a boronic acid derivative of the group Ar with palladium catalysis to form a first intermediate, (2) reacting said first intermediate with a corresponding ortho-substituted organometallic phenyl derivative to form a second intermediate, and (3) reducing said second intermediate.

5. An organic electronic device selected from the group consisting of organic electroluminescent devices, organic field-effect transistors, organic thin-film transistors, organic light-emitting transistors, organic integrated circuits, organic solar cells, organic field-quench devices, organic photo receptors, light-emitting electrochemical cells and organic laser diodes, comprising at least one compound of claim **1**.

6. An organic electroluminescent device comprising at least one compound of claim **1**.

7. The organic electroluminescent device of claim **6**, comprising an anode, a cathode, and at least one emitting layer, and optionally comprising further layers selected from the group consisting of hole-injection layers, hole-transport layers, hole-blocking layers, electron-transport layers, electron-injection layers, and/or charge-generation layers.

8. The organic electroluminescent device of claim **7**, wherein said organic electroluminescent device comprises a host material comprising a host and a dopant wherein said host material comprises the compound of claim **1** and is used as a fluorescent emitter and/or as an electron-transport material and/or as a hole-blocking material.

9. The organic electroluminescent device of claim **8**, wherein said dopants are selected from the group consisting of aromatic anthraceneamines, aromatic anthracenediamines, aromatic pyreneamines, aromatic pyrenediamines, monostyrylamines, distyrylamines, tristyrylamines, tetrastyrylamines, styrylphosphines, styryl ethers, and arylamines.

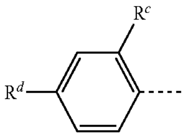
10. The organic electroluminescent device of claim **7**, wherein said compound of claim **1** is used as an emitting compound in an emitting layer and/or as a hole-transport material.

11. The organic electroluminescent device of claim **10**, wherein said hole-transport material is comprised in a hole-transport layer or a hole-injection layer.

12. The organic electroluminescent device of claim **11**, wherein R is an $N(Ar^1)_2$ group and/or R^1 is an $N(Ar^1)_2$ group.

13. The compound of claim **1**, wherein Ar is a group of Formula (9), Formula (10) or Formula (11)

Formula (9)



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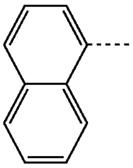
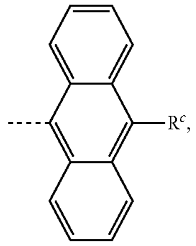
Formula (10)



Formula (11)



wherein R^c and R^d are defined as R^1, with the proviso that R^c in Formula (9) is defined as R.

14. The compound of claim **1**, wherein R is, identically or differently on each occurrence, F, $Si(R^2)_3$, a straight-chain alkyl or alkoxy group having 1 to 4 C atoms or a branched alkyl group having 3 to 5 C atoms, each of which may be substituted by one or more radicals R^2, where one or more H atoms may be replaced by F.

* * * * *

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摘要(译)

本发明涉及蒽衍生物，其在有机电致发光器件中的用途，以及包含这些化合物的有机电致发光器件。

Formula (1)