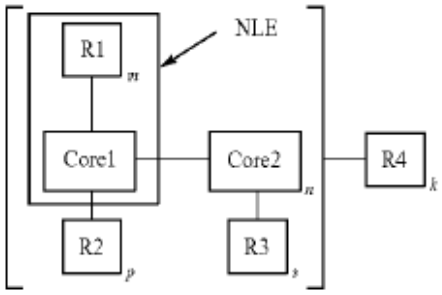
directly or via a connecting group, s is number of the ionic groups R3 which is equal to 0, 1, 2, 3 or 4.

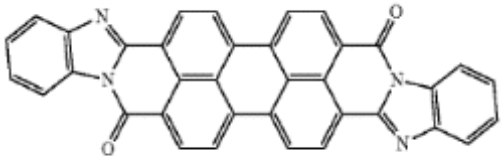
R4 is a resistive substituent providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other and connected to the aromatic polycyclic conjugated molecule (Corel) and/or to the electro-conductive oligomer (Core2) directly or via a connecting group. The parameter k is a number of substituents R4, which is equal to 0, 1, 2, 3, 4, 5, 6, 7 or 8.

In one aspect, the present disclosure refers to an electro-polarizable compound having the following general formula (I):

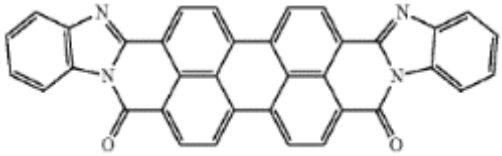


(I)

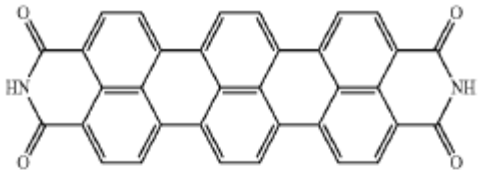
where Corel is an aromatic polycyclic conjugated molecule having two-dimensional flat form and self-assembling by pi-pi stacking in a column-like supramolecule comprising one or more rylene fragments selected from structures 1 to 12:



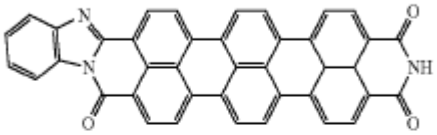
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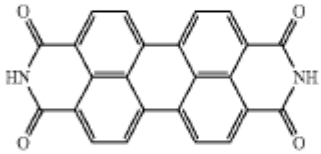
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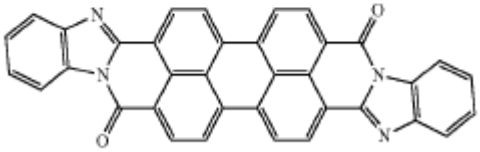
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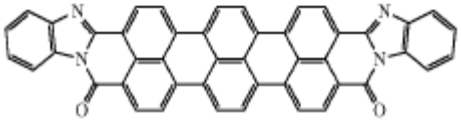
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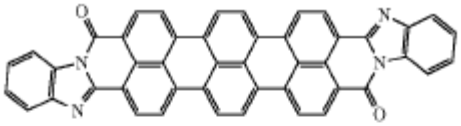
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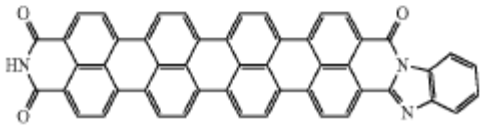
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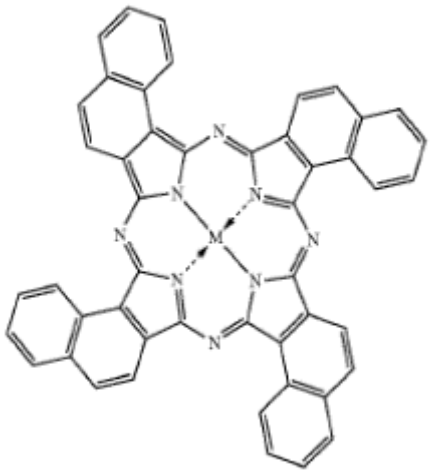


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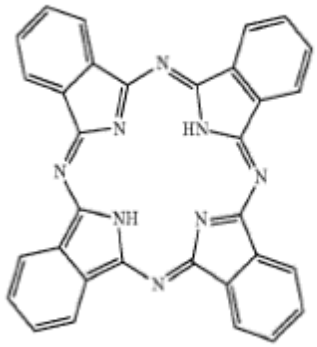
or a tetrapirolic macro-cyclic fragment that has a general structural formula from the group comprising structures 13-19, where M denotes an atom of metal or two protons (2H):



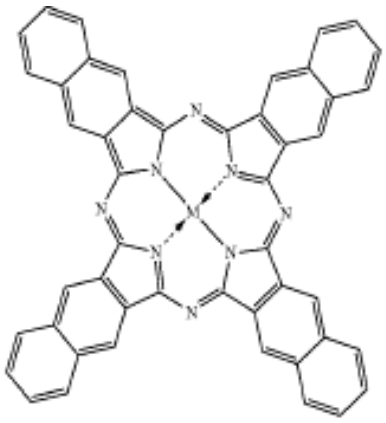
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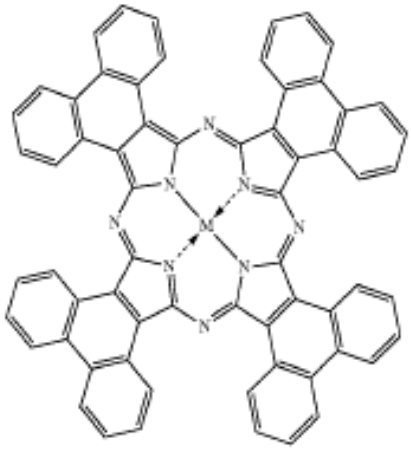
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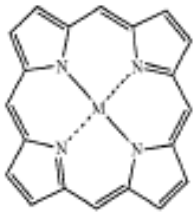
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R1 is an dopant group connected to the aromatic polycyclic conjugated molecule (Corel) selected from nucleophilic groups (donors): —NO_2, --NH_3± and —NR,± (quaternary nitrogen salts), counterion Cl- or Br, —CHO (aldehyde), PRO (keto group), —$S0_3H$ (sulfonic acids), —SO_3R (sulfonates), SO_2NH_2 (sulfo-namides), —COOH (carboxylic acid), —COOR (esters), —00C1 (carboxylic acid chlorides), —$CONH_2$ (amides,), —CF_3, —$CC1_3$, —CN, wherein R is radical selected from the list consisting of alkyl, allyl(—CH_2.—$CHCH_2$), benzyl (—$CH_2C_6H_5$) groups, phenyl, substituted phenyl and other aryl (aromatic) groups and electrophilic groups (acceptors): (phenoxides,), —NH_2, —NHR, NR_2, —OH, OR (ethers), —NHCOR (amides), —OCOR (esters), alkyls, C_6H_5, vinyls, wherein R is radical selected from the list consisting of alkyl, allyl (—CH2-CHCH2) benzyl (—CH2C6H5) groups, phenyl, substituted phenyl and other aryl (aromatic) groups;

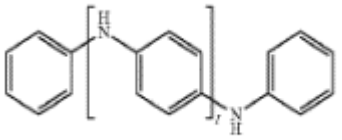
m is number of dopant groups R1 which is equal to 1, 2, 3 or 4;

R2 is a substituent comprising one or more ionic groups from a class of ionic compounds form ionic liquids connected to the aromatic polycyclic conjugated molecule (Corel) directly or via a connecting group;

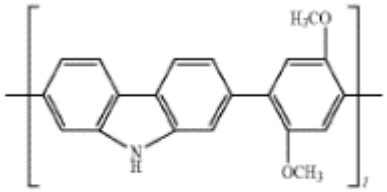
p is number of ionic groups R2 which is equal to 0, 1, 2, 3 or 4;

wherein the fragment marked NLE containing the Corel with at least one dopant group R1 has a nonlinear effect of polarization;

wherein Core2 is an electro-conductive oligomer self-assembling by pi-pi stacking in a column-like supra- molecule selected from the structures 20 to 28 wherein I=2, 3, 4, 5, 6, 7, 8, 9, 10, 11 or 12, Z is O, S or NR5, and R5 is selected from the group consisting of unsubstituted or substituted C_1-C_{18} alkyl, unsubstituted or substituted C_2-C_{18} alkenyl, unsubstituted or substituted C_2-C_{18} alkynyl, and unsubstituted or substituted C_4-C_1, aryl:



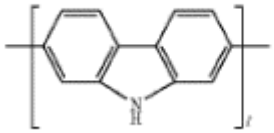
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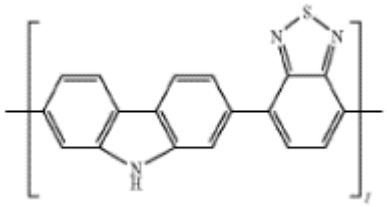
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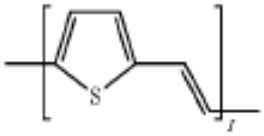
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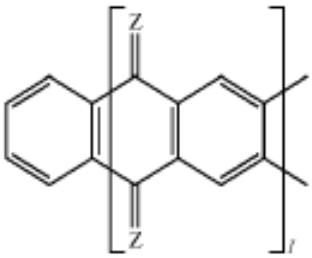
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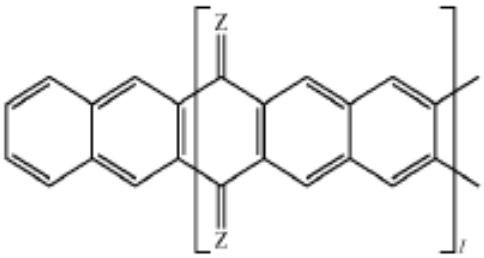
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n is number of the electro-conductive oligomers which is equal to 2, or 4, R3 is a substituent comprising one or more ionic groups from a class of ionic compounds that form ionic liquids connected to the electro-conductive oligomer (Core2) directly or via a connecting group;

s is number of the ionic groups R3 which is equal to 0, 1, 3 or 4;

wherein R4 is a non-polar resistive substituent providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other;

k is a number of substituents R4 which is equal to 0, 1, 2, 4, 5, 6, 7 or 8.

In one example, the present disclosure provides a solution comprising an organic solvent and at least one disclosed electro-polarizable compound.

In another aspect, the present disclosure refers to a solution comprising an organic solvent and at least one type of electro-polarizable compound as disclosed herein.

In another example, the present disclosure provides a crystal metadielectric layer comprising a mixture of the electro-polarizable compounds as disclosed above. The nonlinearly polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one group Rl are placed into the resistive dielectric envelope formed by resistive substituents R4 providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other.

In another aspect, the present disclosure refers to a crystal metadielectric layer comprising a mixture of the electro-polarizable compounds as disclosed herein**,** wherein the nonlinearly polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one dopant group, the electro-conductive oligomers and the ionic groups which have electronic and/or ionic type of polarizability are placed into the resistive dielectric envelope formed by resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other.

In still another example, the present invention provides a meta-capacitor comprising two metal electrodes positioned parallel to each other and which can be rolled or flat and planar with said metadielectric layer between said electrodes, wherein the metadielectric layer comprises one or more types of the disclosed electro-polarizable. The nonlinearly polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one group Rl, the electro-conductive oligomers and the ionic groups which have electronic and/or ionic type of polarizability are placed into the resistive dielectric envelope formed by resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the columnlike supramolecules from each other.

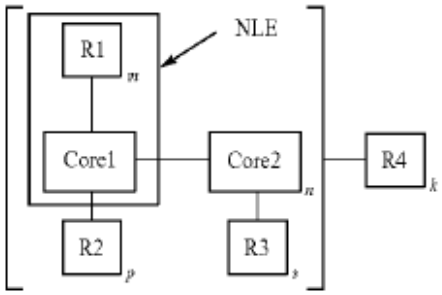
In another aspect, the present disclosure refers to a meta-capacitor comprising two metal electrodes positioned parallel to each other and which can be rolled or flat and planar and metadielectric layer between this electrodes, wherein the layer comprises the electro-polarizable compounds as disclosed herein, wherein the nonlin-early polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one dopant group, the electro-conductive oligomers and the ionic groups which have electronic and/or ionic type of polarizability are placed into the resistive dielectric envelope formed by resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the column-like supra-molecules from each other.

INCORPORATION BY REFERENCE

CLAIMS

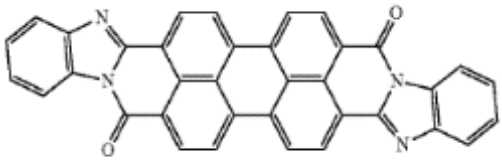
WHAT IS CLAIMED IS:

1. An electro-polarizable compound having the following general formula (I):

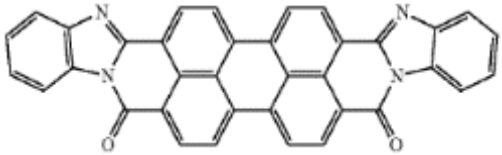


(I)

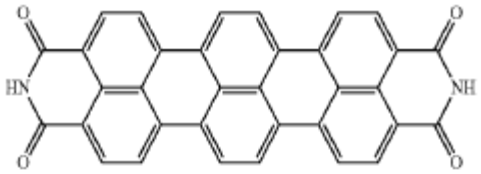
where Corel is an aromatic polycyclic conjugated molecule having two-dimensional flat form and self-assembling by pi-pi stacking in a column-like supramolecule comprising one or more rylene fragments selected from structures 1 to 12:



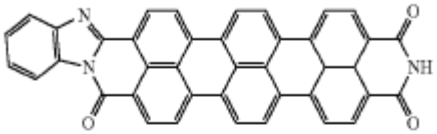
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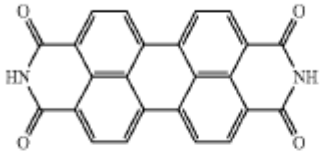
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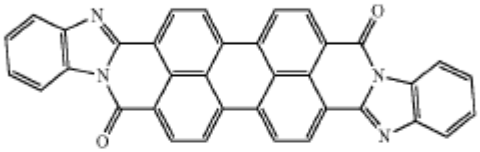
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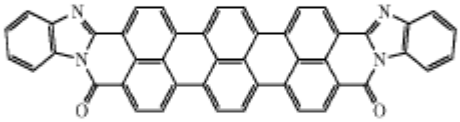
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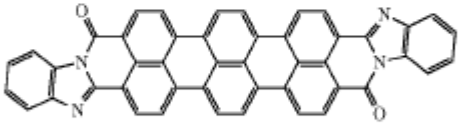
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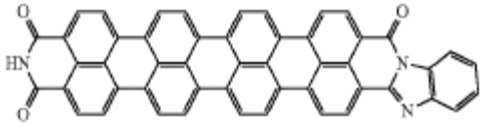
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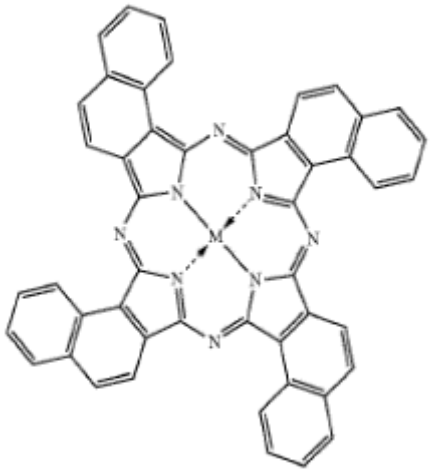


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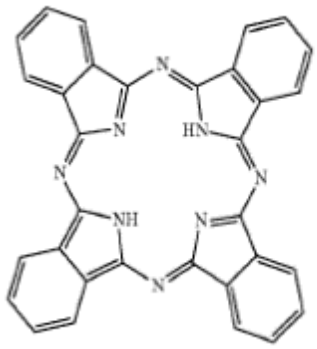
or a tetrapirolic macro-cyclic fragment that has a general structural formula from the group comprising structures 13-19, where M denotes an atom of metal or two protons (2H):



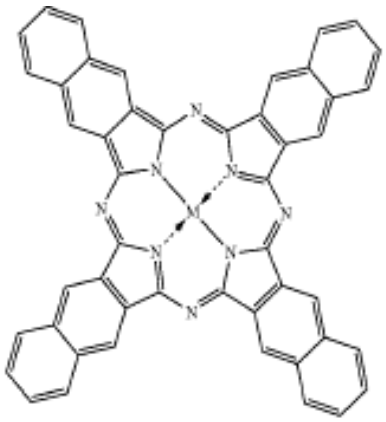
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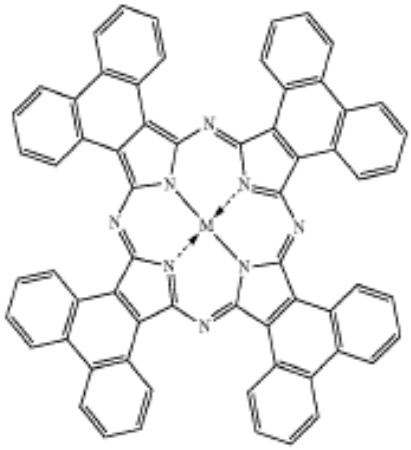
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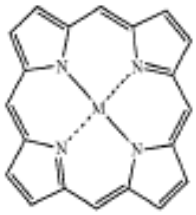
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R1 is an dopant group connected to the aromatic polycyclic conjugated molecule (Corel) selected from nucleophilic groups (donors): —NO_2, --NH_3± and —NR,± (quaternary nitrogen salts), counterion Cl- or Br, —CHO (aldehyde), PRO (keto group), —$S0_3H$ (sulfonic acids), —SO_3R (sulfonates), SO_2NH_2 (sulfo-namides), —COOH (carboxylic acid), —COOR (esters), —00C1 (carboxylic acid chlorides), —$CONH_2$ (amides,), —CF_3, —$CC1_3$, —CN, wherein R is radical selected from the list consisting of alkyl, allyl(—CH_2.—$CHCH_2$), benzyl (—$CH_2C_6H_5$) groups, phenyl, substituted phenyl and other aryl (aromatic) groups and electrophilic groups (acceptors): (phenoxides,), —NH_2, —NHR, NR_2, —OH, OR (ethers), —NHCOR (amides), —OCOR (esters), alkyls, C_6H_5, vinyls, wherein R is radical selected from the list consisting of alkyl, allyl (—CH2-CHCH2) benzyl (—CH2C6H5) groups, phenyl, substituted phenyl and other aryl (aromatic) groups;

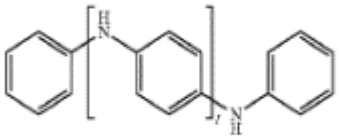
m is number of dopant groups R1 which is equal to 1, 2, 3 or 4;

R2 is a substituent comprising one or more ionic groups from a class of ionic compounds form ionic liquids connected to the aromatic polycyclic conjugated molecule (Corel) directly or via a connecting group;

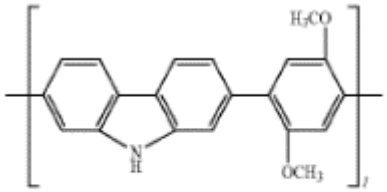
p is number of ionic groups R2 which is equal to 0, 1, 2, 3 or 4;

wherein the fragment marked NLE containing the Corel with at least one dopant group R1 has a nonlinear effect of polarization;

wherein Core2 is an electro-conductive oligomer self-assembling by pi-pi stacking in a column-like supra- molecule selected from the structures 20 to 28 wherein I=2, 3, 4, 5, 6, 7, 8, 9, 10, 11 or 12, Z is O, S or NR5, and R5 is selected from the group consisting of unsubstituted or substituted C_1-C_{18} alkyl, unsubstituted or substituted C_2-C_{18} alkenyl, unsubstituted or substituted C_2-C_{18} alkynyl, and unsubstituted or substituted C_4-C_1, aryl:



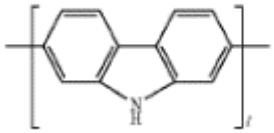
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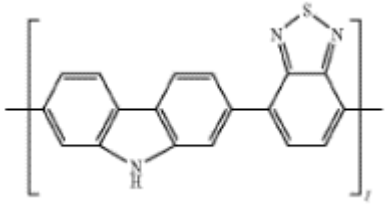
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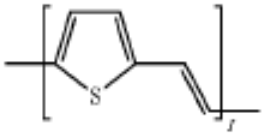
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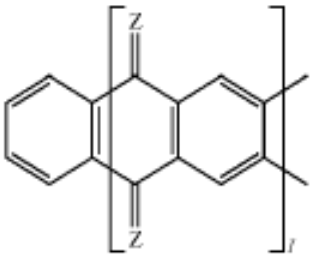
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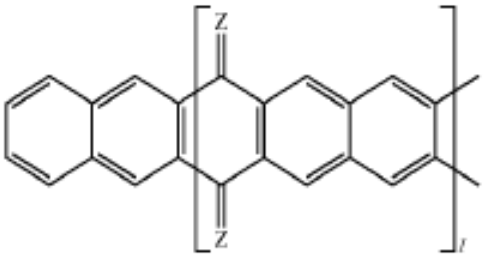
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n is number of the electro-conductive oligomers which is equal to 2, or 4, R3 is a substituent comprising one or more ionic groups from a class of ionic compounds that form ionic liquids connected to the electro-conductive oligomer (Core2) directly or via a connecting group;

s is number of the ionic groups R3 which is equal to 0, 1, 3 or 4;

wherein R4 is a non-polar resistive substituent providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other;

k is a number of substituents R4 which is equal to 0, 1, 2, 4, 5, 6, 7 or 8.

2. The electro-polarizable compound according to claim **1**, wherein at least one ionic group R2 or R3 is independently selected from the list comprising $[NR_4]+$, $[PR_4]+$ as cation and $[—0O_21^-$, $[—S0_3]^-$, $[—P0_3R]^-$, $[—PR_5]^-$ as anion, wherein R is selected from the list comprising H, alkyl, and fluorine.

3. The electro-polarizable compound according to claim **1**, wherein the at least one connecting group is selected from the list comprising the following structures: 29-39, where W is hydrogen (H) or an alkyl group:

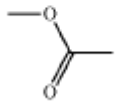
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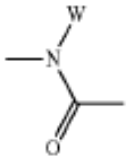


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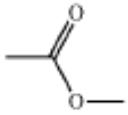


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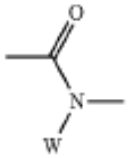




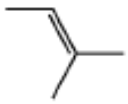
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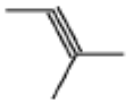
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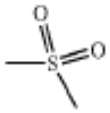
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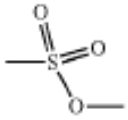
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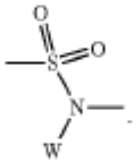
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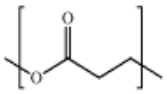
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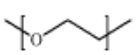


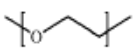
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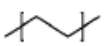
4. The electro-polarizable compound according to claim **1**, wherein the at least one connecting group is selected from the group of CH2, CF2, SiR2O, CH2CH2O, wherein R is 5. selected from the list comprising H, alkyl, and fluorine.

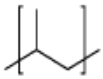
5. The electro-polarizable compound according to claim **1**, wherein the at least one connecting group is selected from structures 40 to 45:


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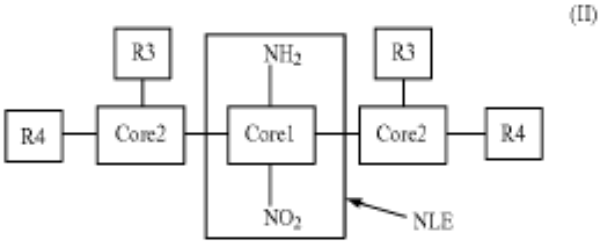
6. The electro-polarizable compound according to claim **1**, wherein the resistive substituent R4 is selected from the group of alkyl, aryl, substituted alkyl, substituted aryl, fluorinated alkyl, chlorinated alkyl, branched and complex alkyl, branched and complex fluorinated alkyl, branched and complex chlorinated alkyl groups, and any combination thereof, and wherein the alkyl group is selected from methyl, ethyl, propyl, n-butyl, iso-butyl and tert-butyl groups, and the aryl group is selected from phenyl, benzyl and naphthyl groups or siloxane, and/or polyethyleneglycol as linear or branched chains.

7. The electro-polarizable compound according to claim **1**, wherein the resistive substituent R4 is $C_{,,Q2x+1,}$ where X≥1 and Q is hydrogen (H), fluorine (F), or chlorine (Cl).

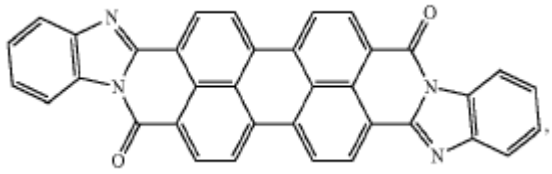
8. The electro-polarizable compound of claim **1**, wherein the aromatic polycyclic conjugated molecule (Corel) and the dopant groups (Rl) form a non-centrosymmetric molecular structure.

9. The electro-polarizable compound of claim **1**, wherein the aromatic polycyclic conjugated molecule (Corel), the dopant groups (Rl) and the resistive substituents (R4) form a non-centrosymmetric molecular structure.

10. A electro-polarizable compound of claim **1** having the following general formula (II):

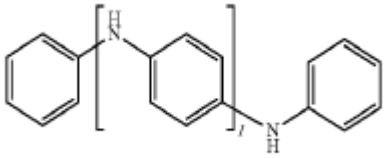


wherein the Corel is rylene fragment having the following structural formula:



wherein two (m is equal to 2) dopant groups —NH_2 and —NO_2 are located on rylene phenyl rings and/or apex 55 phenyl ring positions of the Corel,

wherein the Core2 is the electro-conductive oligomer having the following structural

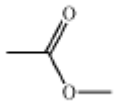


formula:

wherein I=2, 3, 4, 5, 6, 7, 8, 9, 10, 11 or 12, number n of the electro-conductive oligomers is equal to 0 or 2 and the two Core2's are located in apex positions of the Corel,

wherein R3 is the ionic group numbers of the ionic groups R3 is equal to 2,

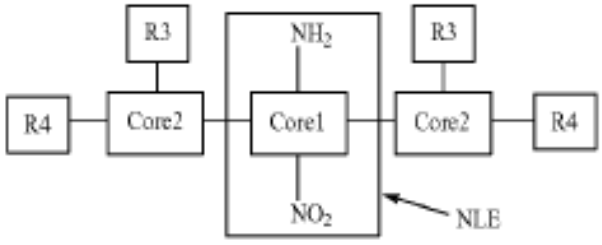
wherein the ionic groups are connected to the electro-conductive oligomer (Core2) via a connecting group having the following structural formula:



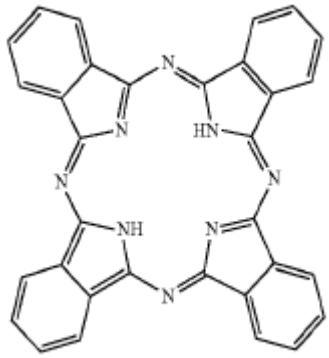
wherein R4 is C_1,14_{37} is a resistive substituent located on the terminus positions of Core2 or apex phenyl rings of Core 1.

11. A electro-polarizable compound of claim 1 having the following general formula:

(III)

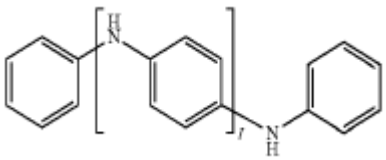


wherein the Corel is tetrapirolic macro-cyclic fragments having the following structural formula:



wherein the Corel comprises two (m is equal to 2) dopant groups —NH_2 and

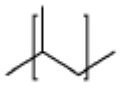
—NO_2 located on opposite apex phenyl ring positions of the Corel, wherein the Core2 is the electro-conductive oligomer having the following structural formula:



wherein I=2, 3, 4, 5, 6, 7, 8, 9, 10, 11 or 12, number n of the electro-conductive oligomers is equal to 0 or 2 and the two Core2 are located in apex positions of the Corel,

wherein R3 is the ionic group $C00^-$, numbers of the ionic groups R3 is equal to 2,

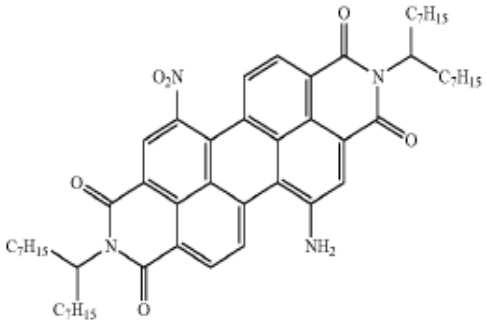
wherein the ionic groups are connected to the electro-conductive oligomer (Core2) via a connecting group having following structural formula:



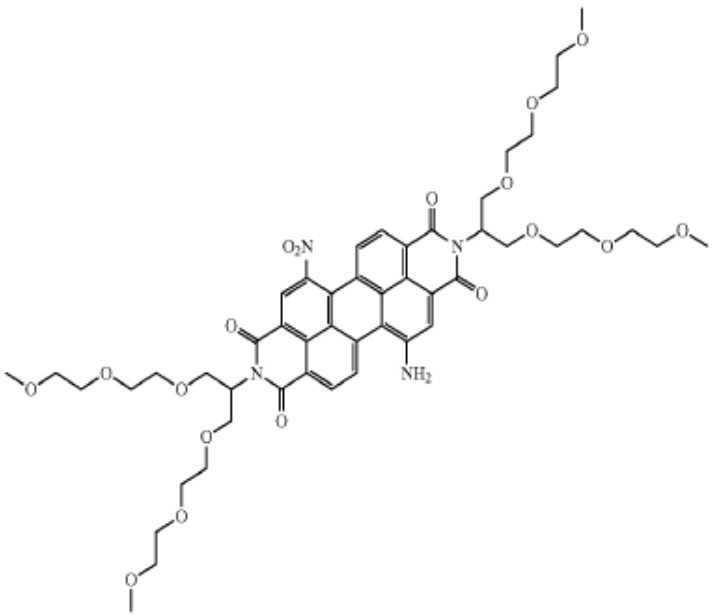
wherein R4 is (C_1-C_{20}) alkyl-resistive substituents located on apex phenyl ring positions of the Core2 groups, or on outer rings of Corel when n is equal to 0.

12. An electro-polarizable compound of claim 1, wherein a fragment comprising the aromatic polycyclic conjugated molecule (Corel), dopant groups (R1) and resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other (R4) is selected from structures 46 to 97:

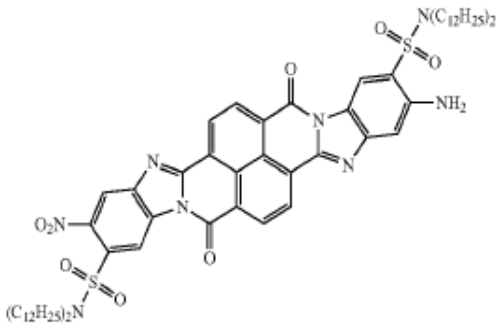
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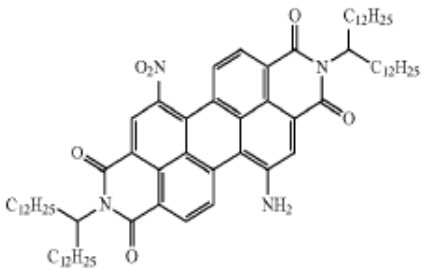
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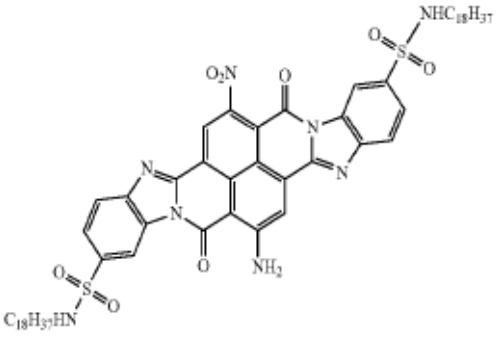
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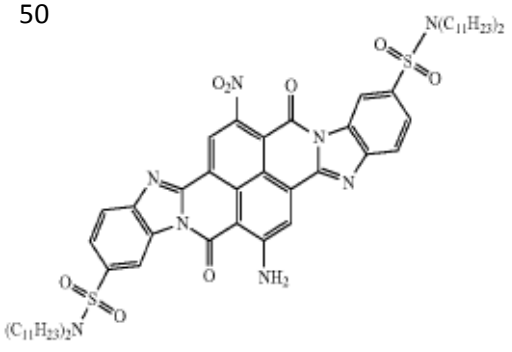
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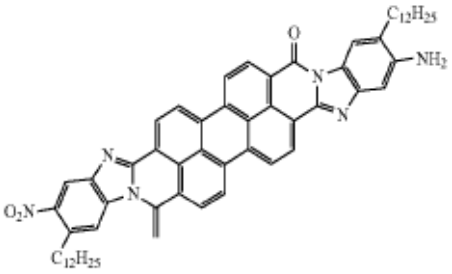
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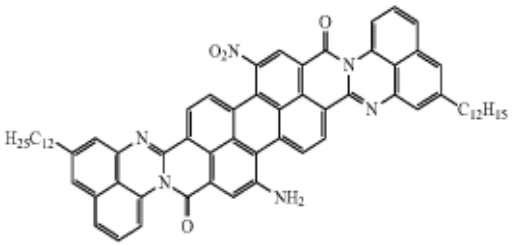
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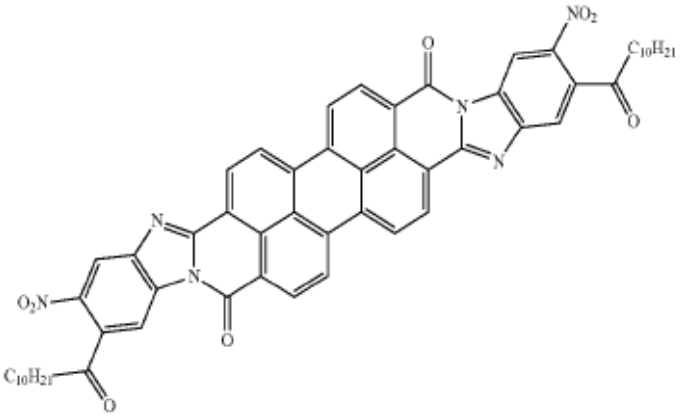
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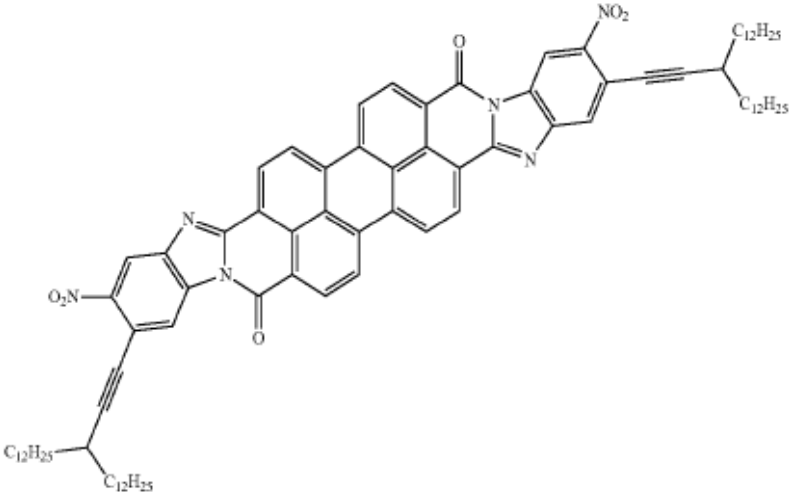
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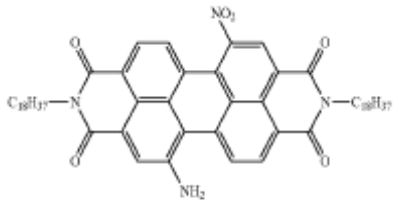


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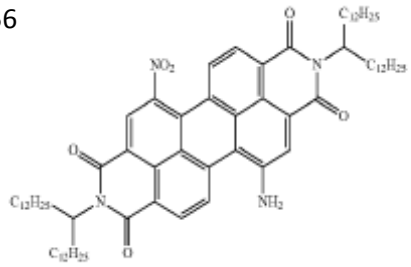


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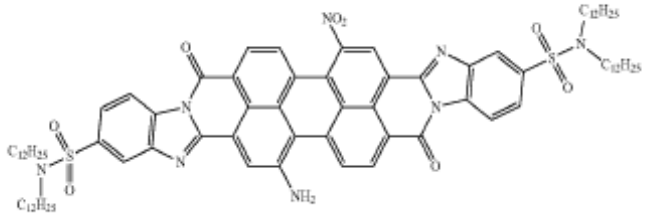
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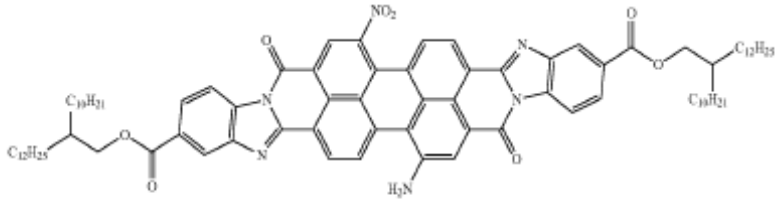
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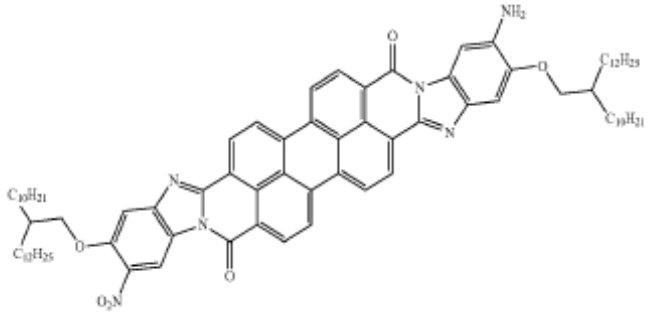
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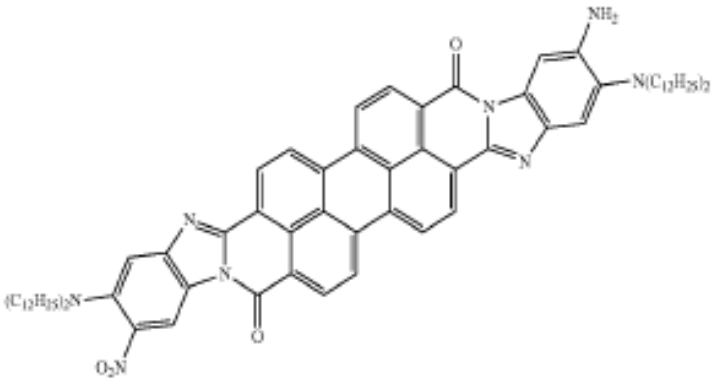


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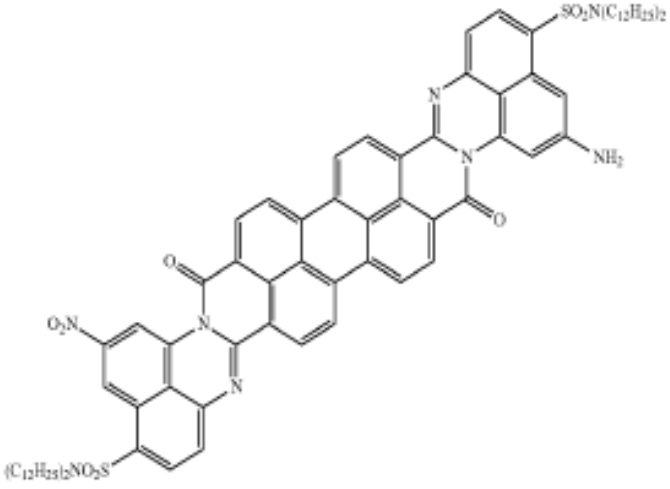


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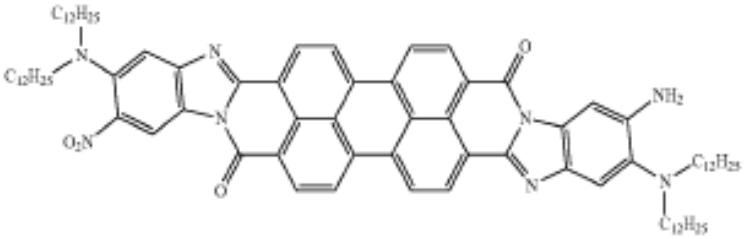
62


NH2
N(C12H25)2
(C12H25)2N
O2N

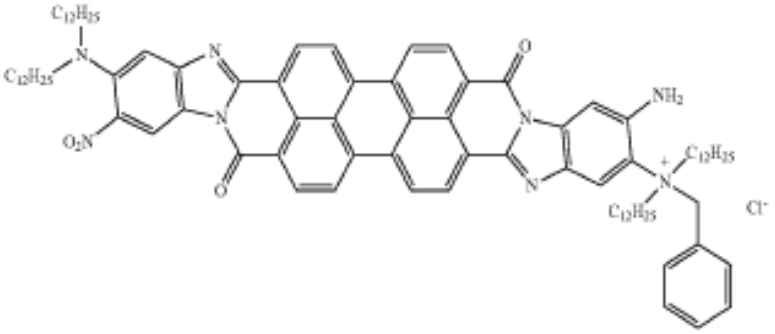
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SO2N(C12H25)2
NH2
O2N
(C12H25)2NO2S

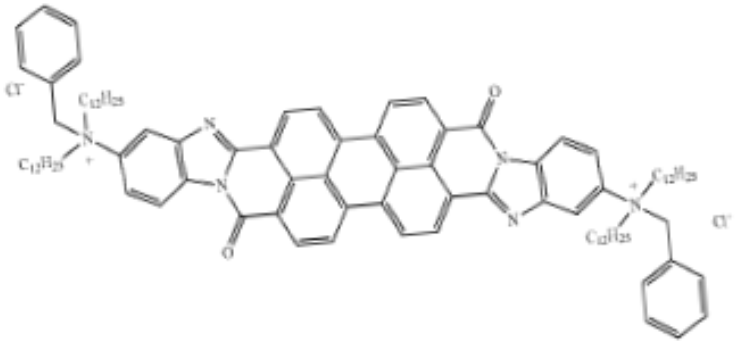
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C12H25
C12H25
O2N
NH2
C12H25
C12H25

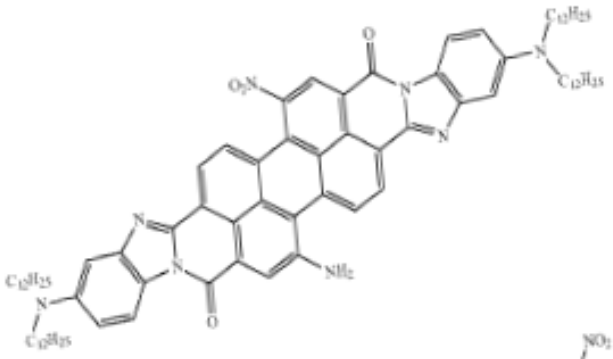
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C12H25
C12H25
O2N
NH2
C12H25
C12H25
Cl-

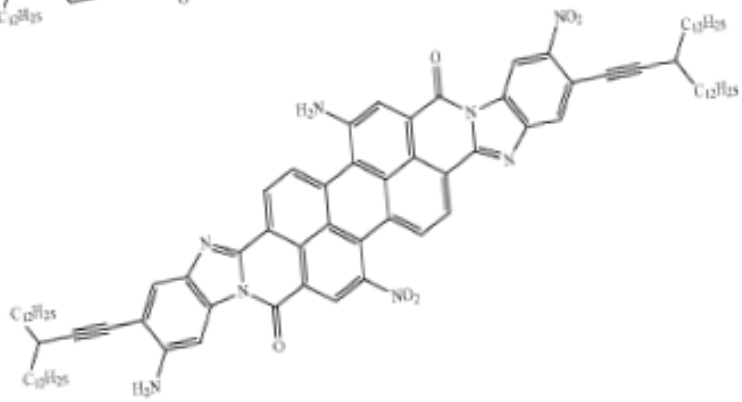
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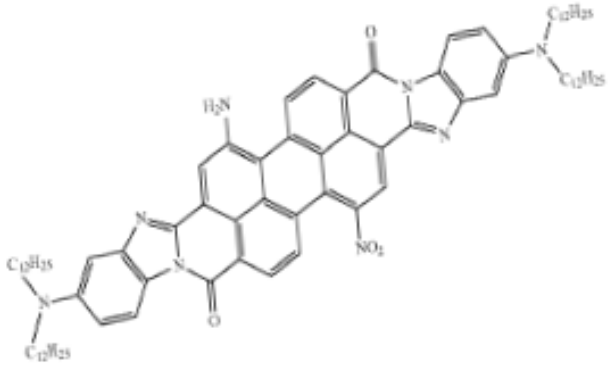
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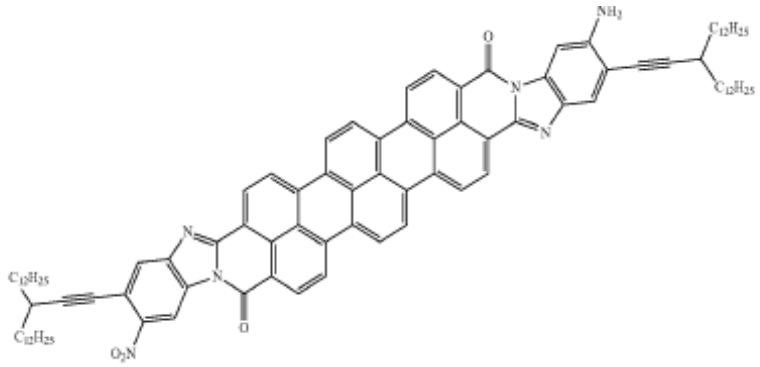


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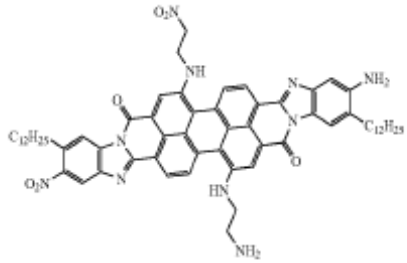
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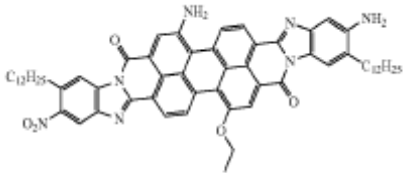




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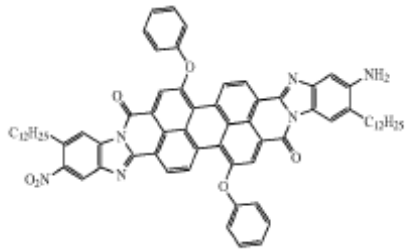
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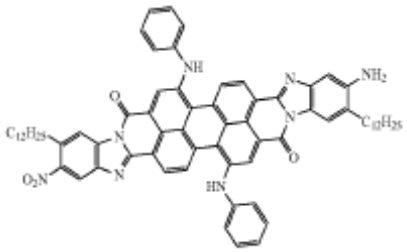




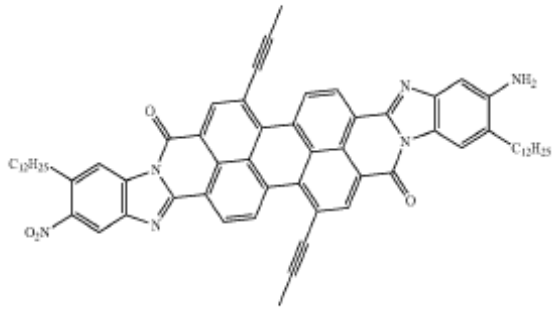
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73



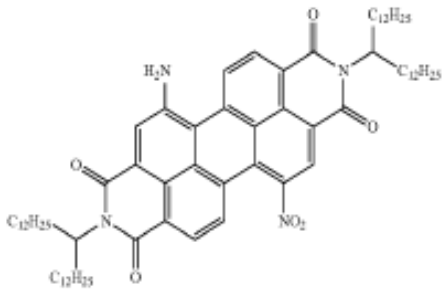


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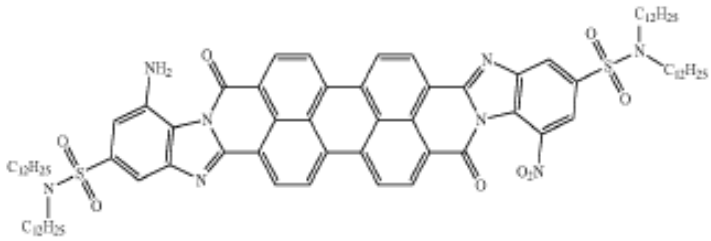


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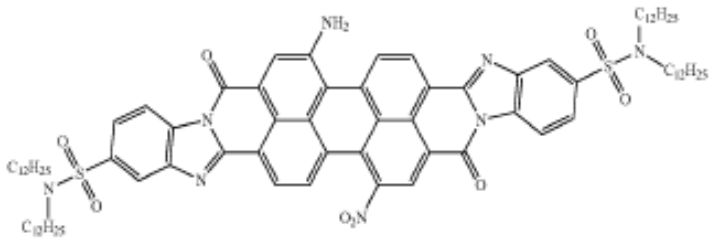
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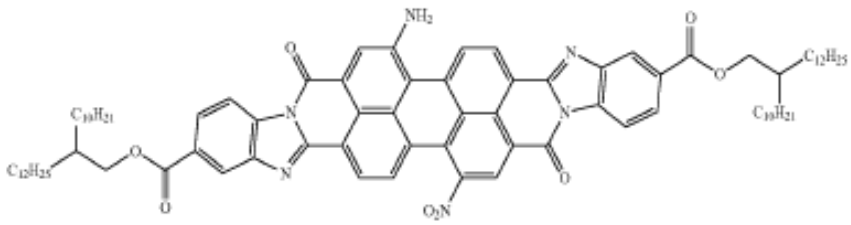
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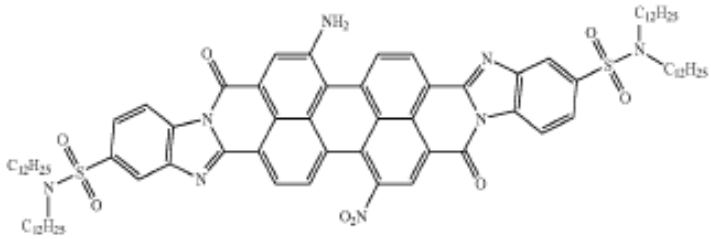
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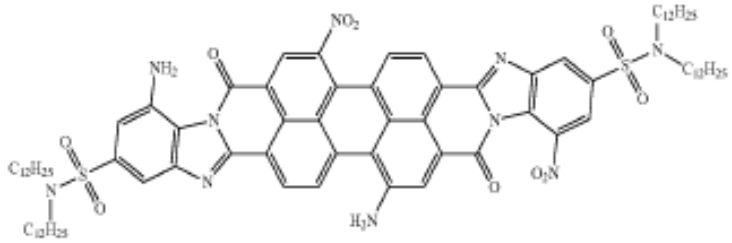
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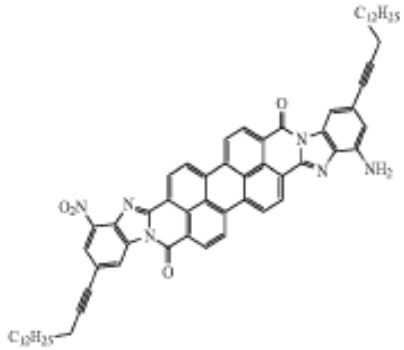
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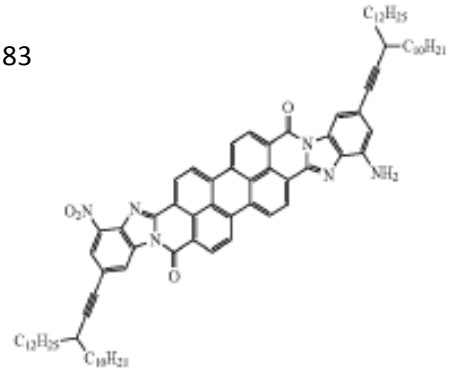
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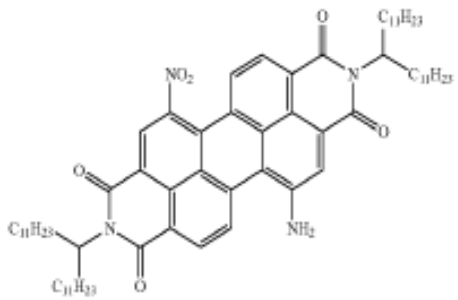
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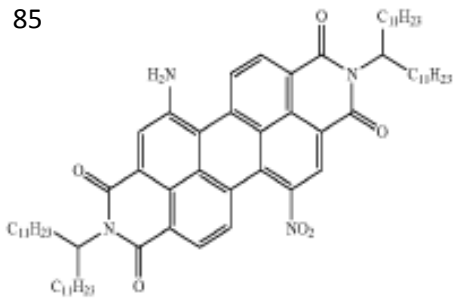
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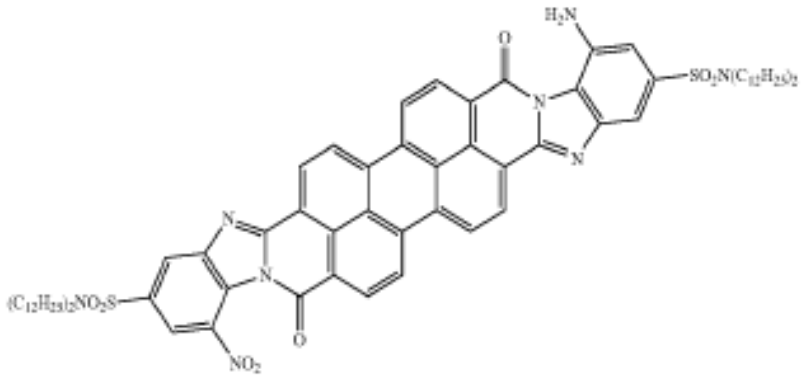
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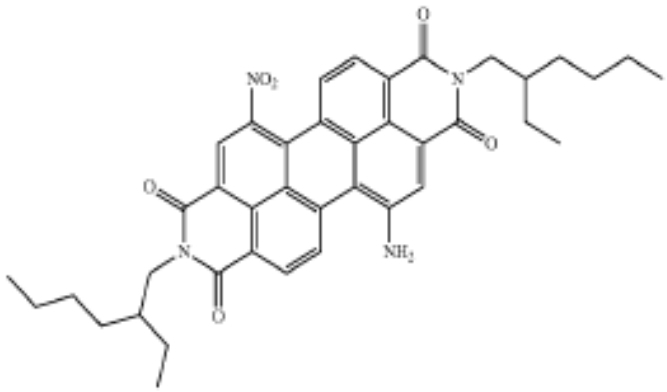
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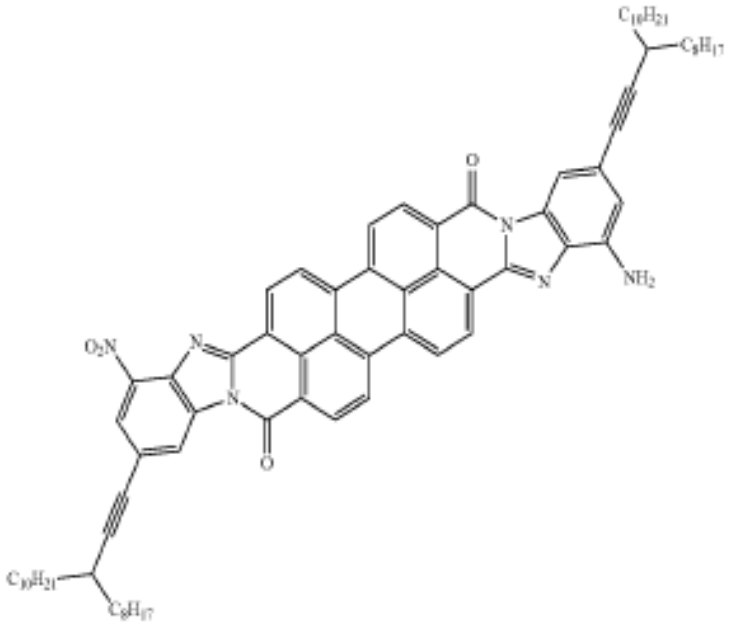
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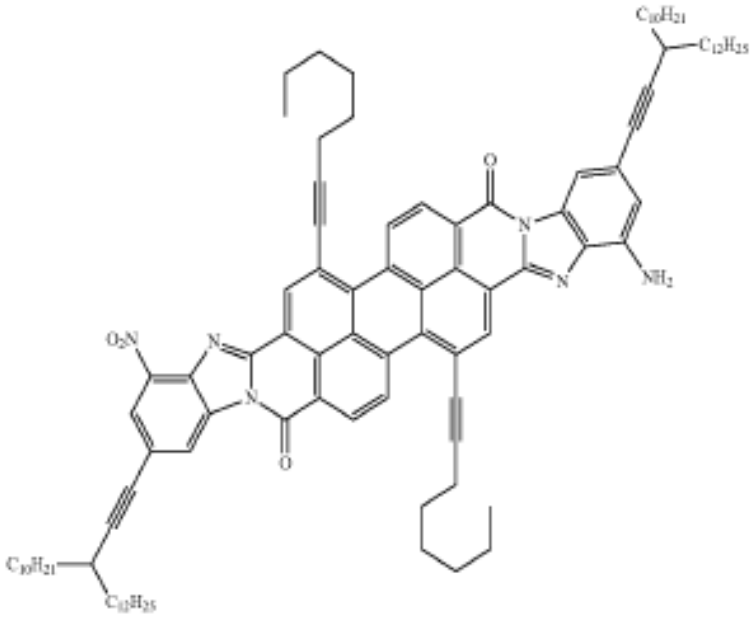
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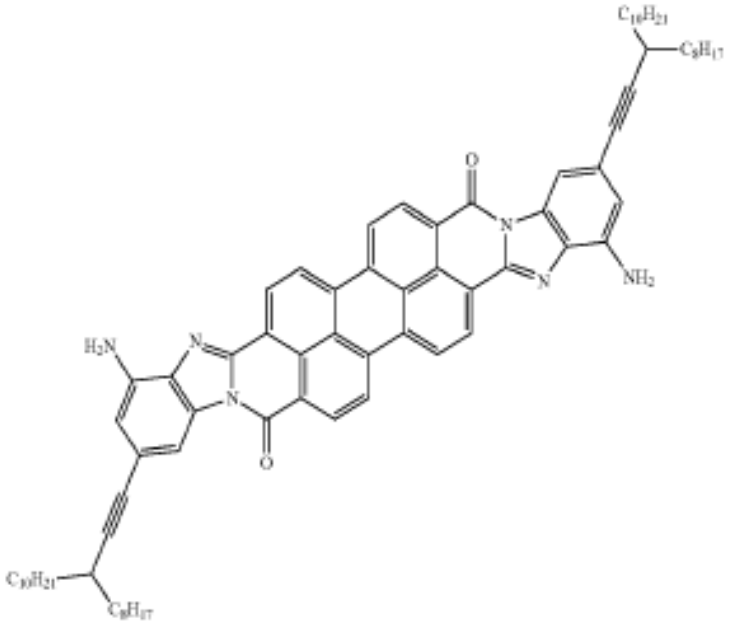
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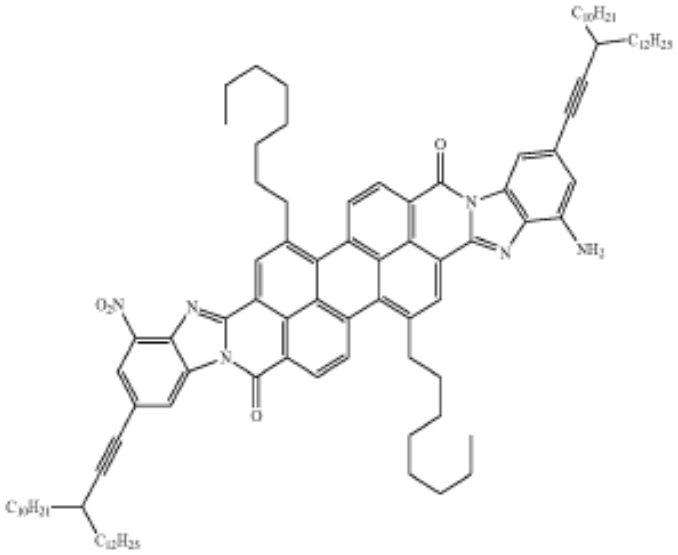
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$C_{10}H_{21}$
$C_{12}H_{25}$
O
N
N
NH_2
O_2N
N
N
O
$C_{10}H_{21}$
$C_{12}H_{25}$

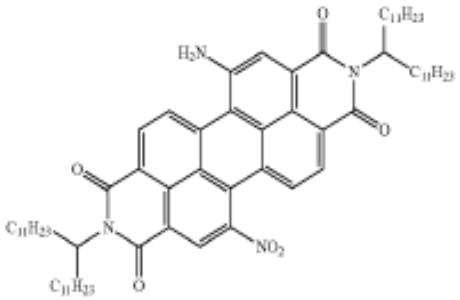
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$C_{10}H_{21}$
C_8H_{17}
O
N
N
NH_2
H_2N
N
N
O
$C_{10}H_{21}$
C_8H_{17}

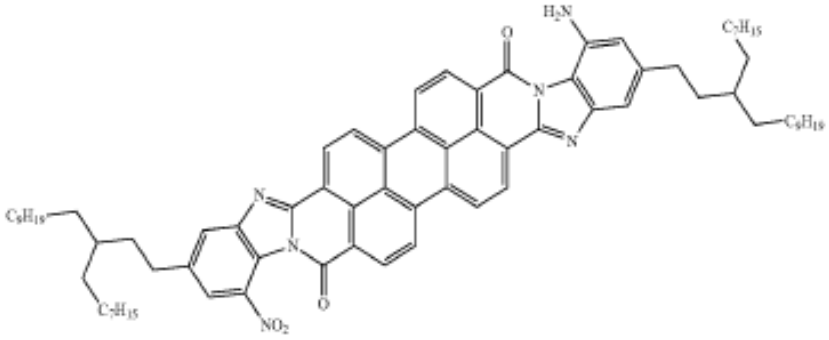
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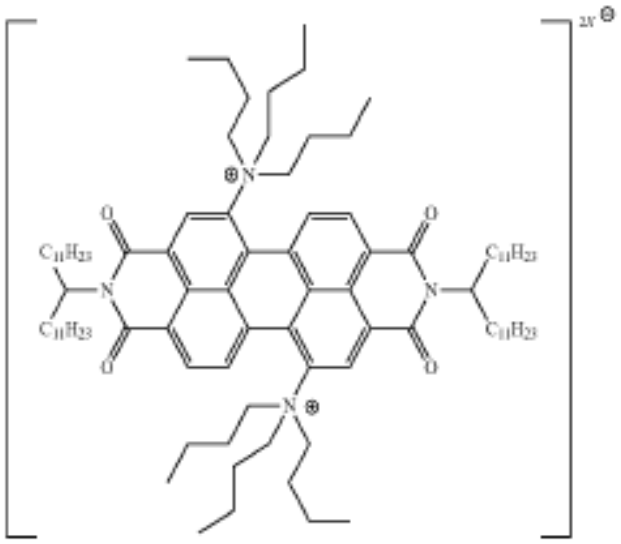


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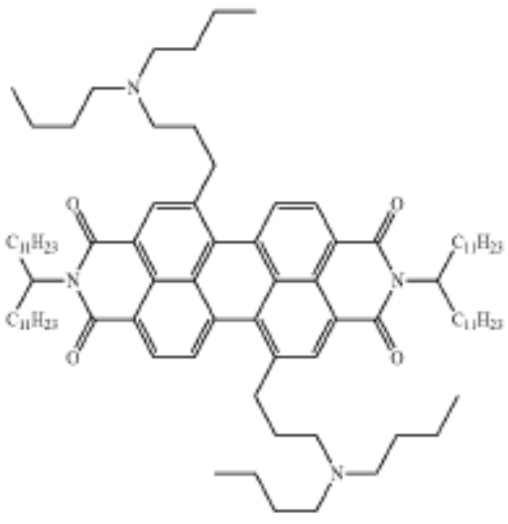
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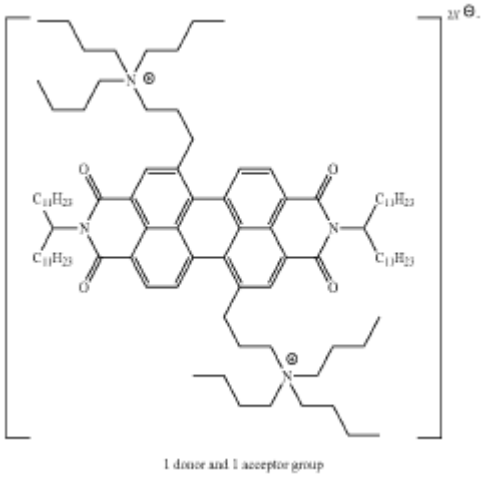


95

One ionic group should be anionic (SO3-, COOH, etc)



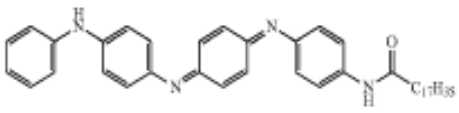
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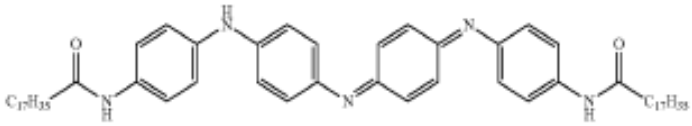
97

13. A electro-polarizable compound of claim **1**, wherein its fragment comprising the electro-conductive oligomer (Core2), resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the column-like supramolecules from each other (R4) and/or the ionic groups R3 is selected from structures 98 to 136:

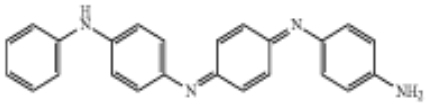
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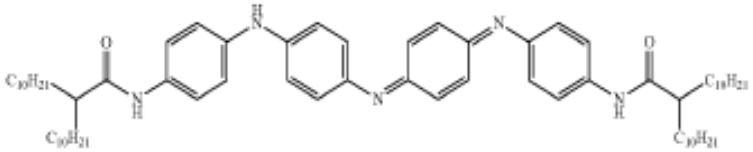
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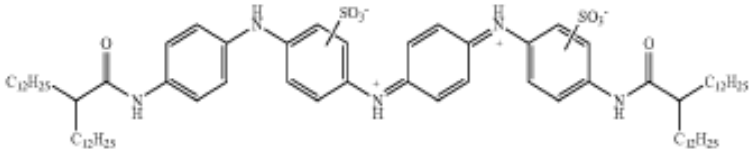
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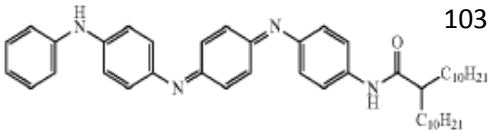


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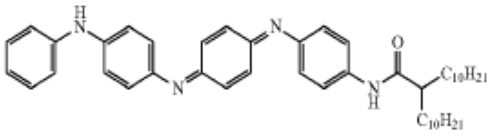


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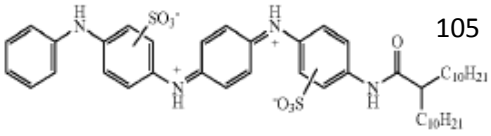




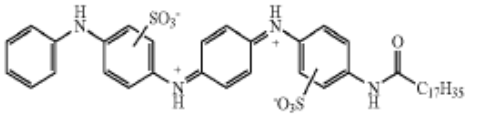
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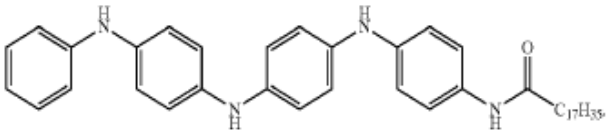
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105



106



107

14. The electro-polarizable compound of claim **1,** wherein the anangement of dopant is chosen such that the electro-polarizable compound has a polarization (a) comprising first-order ($a^{(1)}$) and second-order ($a^{(2)}$) polarization according to follow formula:

$$\alpha=\alpha^{(1)}+\alpha^{(2)}\cdot E,$$

where E is an intensity of external electric field.

15. A solution comprising an organic solvent and at least one type of electro-polarizable compound according to claim **1.**

16. The solution according to claim **15,** comprising a mixture of different electro-polarizable compounds.

17. The solution according to claim **15,** wherein the mixture of the electro-polarizable compounds comprises rylene fragments of different length.

18. The solution according to claim **15,** wherein the organic solvent is selected from the list consisting of ketones, carboxylic acids, hydrocarbons, cyclic hydrocarbons, chlorohydrocarbons, alcohols, ethers, esters, and any combination thereof.

19. The solution according to claim **15,** wherein the organic solvent is selected from the list consisting of acetone, xylene, toluene, ethanol, methylcyclohexane, ethyl acetate, diethyl ether, octane, chloroform, methylene chloride, dichloroethane, trichloroethene, tetrachloroethene, carbon tetrachloride, 1,4-dioxane, tetrahydrofuran, pyridine, triethylamine, nitromethane, acetonitrile, dimethylformamide, dimethyl sulfoxide, and any combination thereof.

20. Any of the solutions according to claim **15,** wherein the solution is a lyotropic liquid crystal solution.

21. A crystal metadielectric layer comprising a mixture of the electro-polarizable compounds according to claim **1,** wherein the nonlinearly polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one dopant group, the electro-conductive oligomers and the ionic groups which have electronic and/or ionic type of polarizability are placed into the resistive dielectric envelope formed by resistive substituents providing solubility of the organic

compound in a solvent and electrically insulating the column-like supramolecules from each other.

22. The crystal metadielectric layer according to claim **21,** wherein the column-like supramolecules are formed by the electro-polarizable compounds comprising rylene fragments of different length.

23. The crystal metadielectric layer according to claim **21,** wherein the layer's relative permittivity is greater than or equal to 1000.

24. The crystal metadielectric layer according to claim **21,** wherein its real part of the relative permittivity (E') comprises first-order (sm) and second-order ($E^{(2)}$) permittivities according to follow formula:

$$\varepsilon' = \varepsilon^{(1)} + 2\varepsilon^{(2)}\frac{V_0}{d},$$

where V_o is the DC-voltage which is applied to the crystal metadielectric layer, d is the layer thickness.

25. The crystal metadielectric layer according to claim **21,** wherein the layer's resistivity is greater than or equal to 10^{13} ohm/cm.

26. A meta-capacitor comprising two metal electrodes positioned parallel to each other and which can be rolled or flat and planar and metadielectric layer between this electrodes, wherein the layer comprises the electro-polarizable compounds according to any claim **1**, wherein the nonlin-early polarizable fragments comprising an aromatic polycyclic conjugated molecule with at least one dopant group, the electro-conductive oligomers and the ionic groups which have electronic and/or ionic type of polarizability are placed into the resistive dielectric envelope formed by resistive substituents providing solubility of the organic compound in a solvent and electrically insulating the column-like supra-molecules from each other.