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(71) Applicant *(for all designated States except US)*: **CIBA SPECIALTY CHEMICALS HOLDING INC.** [CH/CH]; Klybeckstrasse 141, CH-4057 Basel (CH).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: **FRITZSCHE, Katharina** [DE/DE]; Mittlere Strasse 33, 79576 Weil am Rhein (DE). **GROB, Markus** [CH/CH]; Dinkelweg 3, CH-4153 Reinach (CH). **BRAIG, Adalbert** [DE/DE]; Baselstrasse 20, 79589 Binzen (DE). **KIENZLE, Ilona Marion** [DE/DE]; Rheinstrasse 22, 79576 Weil am Rhein/Märkt (DE). **VILAIN, Gérard Daniel Georges** [DE/DE]; Kaiserstuhlstrasse 7, 79285 Ebringen (DE).

(74) Common Representative: CIBA SPECIALTY CHEMICALS HOLDING INC.; Patent Department, Klybeckstrasse 141, CH-4057 Basel (CH).

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(54) Title: LONG WAVELENGTH SHIFTED BENZOTRIAZOLE UV-ABSORBERS AND THEIR USE

(57) Abstract: The instant invention relates to novel benzotriazole UV-absorbers having a long wavelength shifted absorption spectrum with significant absorbance up to 410-420 nm. Further aspects of the invention are a process for their preparation, a UV stabilized composition containing the new UV-absorbers and the use of the new compounds as UV-light stabilizers for organic materials.

Long Wavelength Shifted Benzotriazole UV-Absorbers And Their Use

The instant invention relates to novel benzotriazole UV-absorbers having a long wavelength shifted absorption spectrum with significant absorbance up to 410-430 nm. Further aspects of the invention are a process for their preparation, a UV stabilized composition containing the new UV-absorbers and the use of the new compounds as UV-light stabilizers for organic materials.

Polymeric substrates containing aromatic moieties, such as for example adhesives or coating resins based on aromatic epoxides, aromatic polyesters or aromatic (poly-) isocyanates are highly sensitive to UV/VIS radiation up to wavelengths of approximately 410 to 420 nm.

The protection of such adhesive or coating layers with a UV absorbing layer on top is extremely difficult, since already very small amounts of radiation - even in the range of around 410 nm - penetrating the UV absorbing top coating are sufficient to cause delamination and peeling off of the protective coating.

Typical applications, in which long wavelength shifted UV absorbers are extremely useful, are automotive coatings, typically two coat metallic automotive coatings.

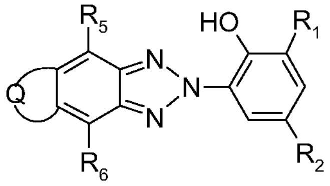
Today’s automotive coatings have applied an anticorrosive cathodic electro coat directly on the steel plate. Due to the significantly red shifted light sensitivity of the cathodic resins (up to approximately 400 – 420nm) it is not possible to protect the cathodic electro coat with conventional prior art UV-absorbers in the top coatings adequately.

In order to better protect such sensitive layers, attempts have been made to shift the UV absorption of benzotriazoles towards longer wavelengths. For example, US 5 436 349 discloses benzotriazole UV-absorbers, which are substituted in the 5 position of the benzo ring by alkylsulfonyl groups. However, the absorption shift is not large enough to protect such highly sensitive materials.

US 6,166,218 discloses, for example, benzotriazole UV-absorbers, which are substituted in the 5-position of the benzo ring with a CF_3 group, leading also to a slightly long wavelength shifted absorbance and to an enhanced photochemical stability. This absorption shift, however, is by far not large enough to protect materials with a photochemical sensitivity up to 410 nm.

Surprisingly it has now been found, that when the benzo ring of benzotriazole UV-absorbers is part of a heterocyclic system a large shift of the absorption maximum of approximately 20 –40 nm is observed, extending up to 430 nm with a steep slope towards the visible region. The compounds show very high extinction coefficients and remain unexpectedly photochemically stable and show virtually no migration in typical coating applications.

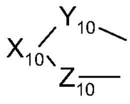
One aspect of the instant invention is a compound of formula (I)



(I)

wherein

Q is a heterocyclic radical forming a 5 or 6-membered ring together with the annealed phenyl-ring, which radical is selected from the group consisting of

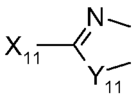


wherein

X₁₀ is O=C, S=C, $R_{101}N$, $S(O)_n$ where n is 0, 1 or 2;

Y₁₀ is NR_{101}, O, $S(O)_n$ where n is 0, 1 or 2;

Z₁₀ is O=C, S=C, $R_{101}N$, O, $S(O)_n$ where n is 0, 1 or 2;



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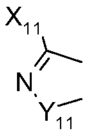
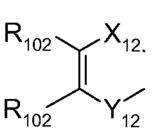


wherein

X₁₁ is R_{101}, $N(R_{101})_2$, OR_{101}, $S(O)_nR_{101}$ where n is 0, 1 or 2;

Y₁₁ is C=O, NR_{101}, O, or $S(O)_n$ where n is 0, 1 or 2;



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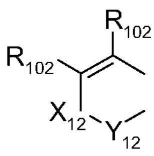

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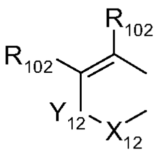

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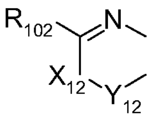

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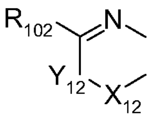

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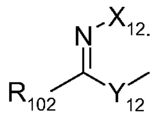
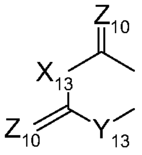


wherein

X_{12} is C=O or C=S;

Y_{12} is NR_{101}, O or S;



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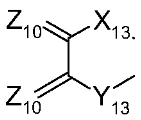


wherein

X_{13} and **Y_{13}** are independently NR_{101}, O or S;

the **Z_{10}** are independently from each other O or S;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more -halogen, -OH, -OR_{22}, -NH_2, -NHR_{22}, -$N(R_{22})_2$, -$NHCOR_{23}$, -$NR_{22}COR_{23}$, -$OCOR_{24}$, -COR_{25}, -SO_2R_{26}, -$SO_3^-M^+$, -$PO(R_{27})_n(R_{28})_{2-n}$, -$Si(R_{29})_n(R_{30})_{3-n}$, -$Si(R_{22})_3$, -$N^+(R_{22})_3$ A^-, -$S^+(R_{22})_2$ A^- or combinations thereof; said straight or branched chain unsubstituted or substituted C_1-C_{24} alkyl, straight or branched chain unsubstituted or substituted C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

said phenyl, naphthyl or C_7-C_{15}phenylalkyl can also be substituted by one ore more -halogen, -CN, -CF_3, -NO_2, -NHR_{22}, -$N(R_{22})_2$, -SO_2R_{26}, -$PO(R_{27})_n(R_{28})_{2-n}$, -OH, -$OR_{22}$, -$COR_{25}$, -$R_{25}$; wherein

n is 0, 1 or 2;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl, C_7-C_{15} phenylalkyl, or two R_{22} when attached to an N or Si atom can form together with the atom to which they are bonded a pyrrolidine, piperidine or morpholine ring;

R_{23} is hydrogen, OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{25} is hydrogen, OH, OR_{22}, NHR_{22} or $N(R_{22})_2$, or has the same meaning as R_{22},

R_{26} is OH, OR_{22}, NHR_{22} or $N(R_{22})_2$,

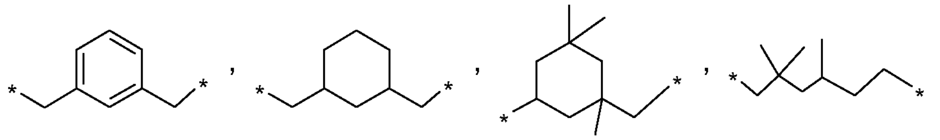
R_{27} is NH_2, NHR_{22} or $N(R_{22})_2$,

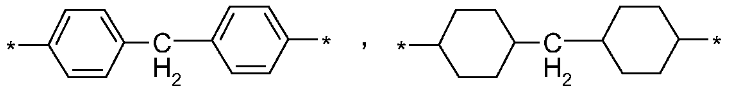
R_{28} is OH or OR_{22},

R_{29} is Cl or OR_{22},

R_{30} is straight or branched chain C_1-C_{18} alkyl; or

R_{101} can be a bridging group of straight or branched C_1-C_{18}alkylene, C_5-C_{10}cycloalkylene, para-phenylene or a group





wherein * denotes a bond where the bridge connects two compounds of formula I, said C_1-C_{12}alkylene, C_5-C_{10}cycloalkylene can also be interrupted by one or more -O-, -S-, -NH- or $-NR_{22}-$ groups or combinations thereof;

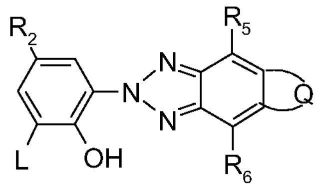
or when Y is a direct bond, Z can additionally also be a direct bond;

R_{102} is hydrogen, -CN, $-COR_{24}$ straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or

if two substituents R_{101}/R_{101}, R_{101}/R_{102} or R_{102}/R_{102} are in vicinal position, they can form together with the atoms to which they are bonded an aliphatic, 5 to 8-membered ring;

R_1 is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms; or

R_1 is a group



wherein

L is alkylene of 1 to 12 carbon atoms, alkylidene of 2 to 12 carbon atoms, benzylidene, p-xylylene or cycloalkylene of 5 to 7 carbon atoms;

R_2 is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

said alkyl substituted by one or more -OH, -OCO-R_{11}, -OR_{14}, -NCO or -NH_2 groups or mixtures thereof, or said alkyl or said alkenyl interrupted by one or more -O-, -NH- or –NR_{14}- groups or mixtures thereof and which can be unsubstituted or substituted by one or more -OH, -OR_{14} or -NH_2 groups or mixtures thereof; where

R_{11} is hydrogen, straight or branched chain C_1-C_{18}alkyl, C_5-C_{12}cycloalkyl, straight or branched chain C_3-C_8alkenyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; and

R_{14} is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms; or

R_2 is –OR_{14}, a group –C(O)-O-R_{14}, -C(O)-NHR_{14} or –C(O)-NR_{14}R'$_{14}$ wherein R'$_{14}$ has the same meaning as R_{14}; or

R_2 is –SR_{13}, -NHR_{13} or -N(R_{13})$_2$; or

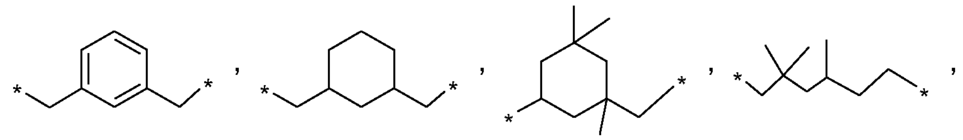
R_2 is -(CH_2)$_m$-CO-X_1-(Z)$_p$-Y-R_{15} wherein

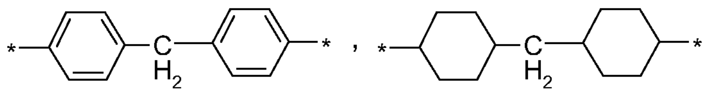
X_1 is -O- or -N(R_{16})-,

Y is -O- or -N(R_{17})- or a direct bond,

Z is C_2-C_{12}-alkylene, C_4-C_{12}alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or is C_3-C_{12}alkylene, butenylene, butynylene, cyclohexylene or phenylene, each of which may be additionally substituted by a hydroxyl group;

or a group





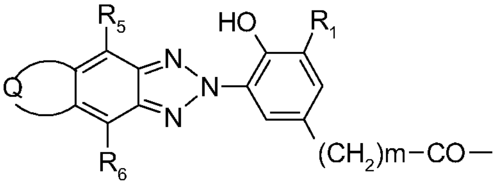
wherein * denotes a bond

or when Y is a direct bond, Z can additionally also be a direct bond;

m is zero, 1 or 2,

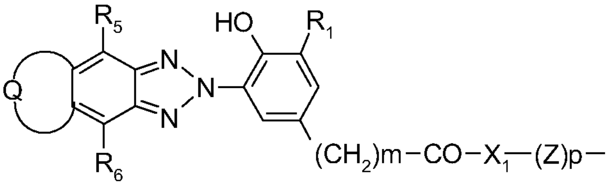
p is 1, or p is also zero when X and Y are -N(R_{16})- and -N(R_{17})-, respectively,

R_{15} is hydrogen, C_1-C_{12}alkyl, a group

 or a group -CO-C(R_{18})=C(H)R_{19} or, when Y is -N(R_{17})-, forms together with R_{17} a group -CO-CH=CH-CO- wherein

R_{18} is hydrogen or methyl, and **R_{19}** is hydrogen, methyl or -CO-X_1-R_{20}, wherein

R_{20} is hydrogen, C_1-C_{12}alkyl or a group of formulae

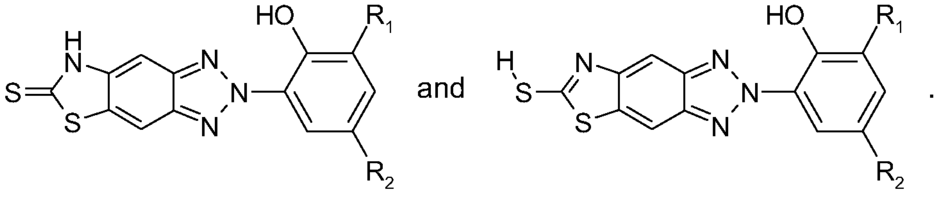
 ;

R_5 and R_6 are independently hydrogen or C_1-C_{18}alkylene;

R_{13} is alkyl of 1 to 20 carbon atoms, hydroxyalkyl of 2 to 20 carbon atoms, alkenyl of 3 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl or naphthyl, which both may be substituted by one or two alkyl of 1 to 4 carbon atoms;

R_{16} and **R_{17}** independently of one another are hydrogen, C_1-C_{12}-alkyl, C_3-C_{12}-alkyl interrupted by 1 to 3 oxygen atoms, or is cyclohexyl or C_7-C_{15}phenylalkyl, and R_{16} together with R_{17} in the case where Z is ethylene, also forms ethylene.

In some cases tautomeric forms may exist. It is intended in the present invention to cover both tautomeric forms, although only one is explicitly outlined. An example is:



When any of the substituents are straight or branched chain alkyl of 1 to 24 carbon atoms, such groups are, for example, methyl, ethyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, tert-amyl, 2-ethylhexyl, tert-octyl, lauryl, tert-dodecyl, tridecyl, n-hexadecyl, n-octadecyl or eicosyl.

When any of said substituents are straight or branched chain alkenyl of 2 to 18 carbon atoms, such groups are, for example, allyl, pentenyl, hexenyl, doceneyl or oleyl.
Preference is given to alkenyl having from 3 to 16, especially from 3 to 12, for example from 2 to 6, carbon atoms.

When any of said substituents are cycloalkyl of 5 to 12 carbon atoms, such groups are, for example, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl and cyclododecyl.
C_1-C_4alkyl-substituted C_5-C_8cycloalkyl is, for example, methylcyclopentyl, dimethylcyclopentyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl or tert-butylcyclohexyl.

When any of said radicals are phenylalkyl of 7 to 15 carbon atoms, such groups are, for example, benzyl, phenethyl, α-methylbenzyl or α,α-dimethylbenzyl.

When phenyl is substituted by alkyl, this is, for example, tolyl and xylyl.

Alkyl substituted by one or more -O- groups and/or substituted by one or more –OH can, for example, be be $-(OCH_2CH_2)_wOH$ or $-(OCH_2CH_2)_wO(C_1\text{-}C_{24}\text{alkyl})$ where w is 1 to 12.

Alkyl interrupted by one or more –O- can be derived from ethylenoxide units or from propyleneoxide units or from mixtures of both.

When alkyl is interrupted by –NH- or $-NR_{14}-$ the radicals are derived in analogy to the above –O- interrupted radicals. Preferred are repeating units of ethylenediamine.

Examples are CH_3-O-CH_2CH_2-, CH_3-NH-CH_2CH_2-, CH_3-N(CH_3)-CH_2-, CH_3-O-CH_2CH_2-O-CH_2CH_2-, CH_3-(O-CH_2CH_2-$)_2$O-CH_2CH_2-, CH_3-(O-CH_2CH_2-$)_3$O-CH_2CH_2- or CH_3-(O-CH_2CH_2-$)_4$O-CH_2CH_2-.

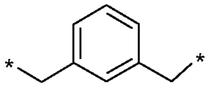
Alkylene is, for example, ethylene, tetramethylene, hexamethylene, 2-methyl-1,4-tetramethylene, hexamethylene, octamethylene, decamethylene and dodecamethylene.

Cycloalkylene is, for example, cyclopentylene, cyclohexylene, cycloheptylene, cyclooctylene and cyclododecylene. Preference is given to cyclohexylene.

Alkylene interrupted by oxygen, NH or $-NR_{14}-$ is, for example, $-CH_2CH_2$-O-CH_2CH_2-,

$-CH_2CH_2-NH-CH_2CH_2-$, $-CH_2CH_2-N(CH_3)-CH_2CH_2-$, $-CH_2CH_2-O-CH_2CH_2-O-CH_2CH_2-$, $-CH_2CH_2-(O-CH_2CH_2-)_2O-CH_2CH_2-$, $-CH_2CH_2-(O-CH_2CH_2-)_3O-CH_2CH_2-$, $-CH_2CH_2-(O-CH_2CH_2-)_4O-CH_2CH_2-$ or $-CH_2CH_2-NH-CH_2CH_2-$.

The radical Q is straight or branched C_1-C_{12}alkylene, C_5-C_{10}cycloalkylene, para-phenylene or a group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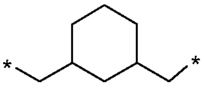
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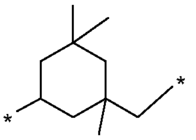
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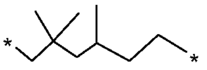
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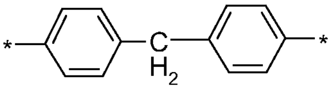
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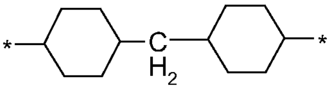
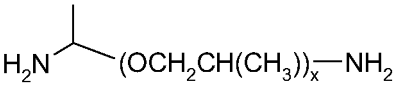

wherein * denotes a bond.

The radical can be derived from readily available diamines, for example, so called Jeffamines. Examples for diamines are Ethylenediamine, propylenediamine, 2-methyl-1,5-pentamethylendiamine, isophorondiamine or 1,2-diaminocyclohexane.

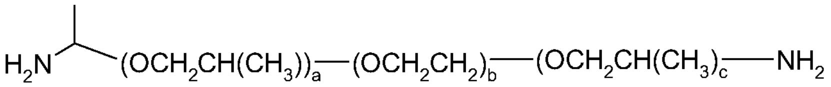
In analogy the radical Z can also be derived from the same available diamines or from the corresponding diols.

Typical Jeffamines are, for example D-2000



$$H_2N-CH(CH_3)-(OCH_2CH(CH_3))_x-NH_2$$

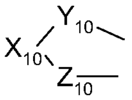
wherein x is 33.1 or ED-2003



$$H_2N-CH(CH_3)-(OCH_2CH(CH_3))_a-(OCH_2CH_2)_b-(OCH_2CH(CH_3))_c-NH_2$$

wherein a + c is 5 and b is 39.5.

For example in the compound of formula (I) Q is



Y_{10} is NR_{101}

X_{10} is O=C, S=C, $R_{101}N$;

Z_{10} is O=C, S=C, $R_{101}N$, O, $S(O)_n$ where n is 0;

or **Y_{10}** is O

X_{10} is O=C, S=C;

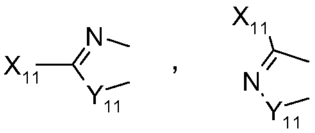
Z_{10} is $R_{101}N$;

or **Y_{10}** is S

X_{10} is O=C, S=C;

Z_{10} is $R_{101}N$;

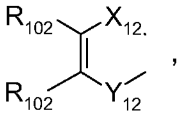
or Q is



X_{11} is R_{101}, $N(R_{101})_2$, OR_{101}, $S(O)_nR_{101}$ where n is 0;

Y_{11} is C=O, NR_{101}, O, or $S(O)_n$ where n is 0;

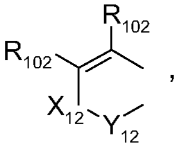
or Q is



X_{12} is C=O;

Y_{12} is NR_{101}, O or S;

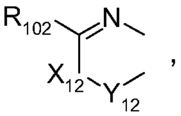
or Q is



X_{12} is C=O;

Y_{12} is NR_{101}, O or S;

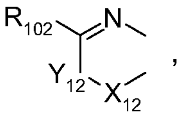
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

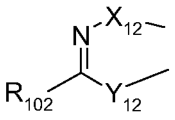
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

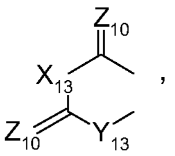
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

or Q is

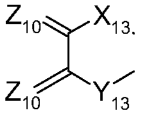


X_{13} is NR_{101}, O or S;

Y_{13} is NR_{101};

Z_{10} is O;

or Q is



X_{13} is NR_{101};

Y_{13} is NR_{101};

Z_{10} is O;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more -halogen, -OH, -OR_{22}, -NH_2, -NHR_{22}, -$N(R_{22})_2$, -$NHCOR_{23}$, -$NR_{22}COR_{23}$, -$OCOR_{24}$, -COR_{25}, -SO_2R_{26}, -$SO_3^-M^+$, -$PO(R_{27})_n(R_{28})_{2-n}$, -$Si(R_{29})_n(R_{30})_{3-n}$, -$Si(R_{22})_3$, -$N^+(R_{22})_3$ A^-, -$S^+(R_{22})_2$ A^- or combinations thereof; said straight or branched chain unsubstituted or substituted C_1-

C_{24} alkyl, straight or branched chain unsubstituted or substituted C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

said phenyl, naphthyl or C_7-C_{15}phenylalkyl can also be substituted by one ore more -halogen, -CN, -CF_3, -NO_2, -NHR_{22}, -$N(R_{22})_2$, -SO_2R_{26}, -$PO(R_{27})_n(R_{28})_{2-n}$, -OH, -$OR_{22}$, -$COR_{25}$, -$R_{25}$; wherein

n is 0, 1 or 2;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl, C_7-C_{15} phenylalkyl, or two R_{22} when attached to an N or Si atom can form together with the atom to which they are bonded a pyrrolidine, piperidine or morpholine ring;

R_{23} is hydrogen, OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{25} is hydrogen, OH, OR_{22}, NHR_{22} or $N(R_{22})_2$, or has the same meaning as R_{22},

R_{26} is OH, OR_{22}, NHR_{22} or $N(R_{22})_2$,

R_{27} is NH_2, NHR_{22} or $N(R_{22})_2$,

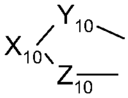
R_{28} is OH or OR_{22},

R_{29} is Cl or OR_{22},

R_{30} is straight or branched chain C_1-C_{18} alkyl; and

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl.

Preferred is a compound of formula (I) wherein Q is



Y_{10} is NR_{101}

X_{10} is O=C, S=C

Z_{10} is O=C, R_{101}N, O, $S(O)_n$ where n is 0;

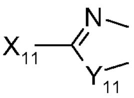
or

Y_{10} is NR_{101}

X_{10} is NR_{101}

Z_{10} is O=C;

or Q is



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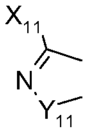
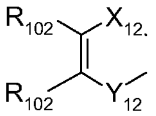


X_{11} is R_{101};

Y_{11} is C=O, NR_{101}, O, or $S(O)_n$ where n is 0;

or Q is

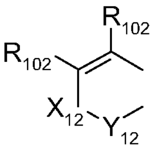


,

X_{12} is C=O;

Y_{12} is NR_{101} or O;

or Q is

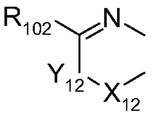


,

X_{12} is C=O;

Y_{12} is NR_{101} or O;

or Q is

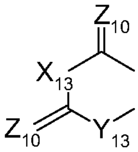


,

X_{12} is C=O;

Y_{12} is NR_{101} or O;

or Q is

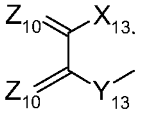


,

X_{13} is NR_{101};

Y_{13} is NR_{101};

Z_{10} is O;

or Q is 

X_{13} is NR_{101};

Y_{13} is NR_{101};

Z_{10} is O;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; said straight or branched chain said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more –OH; or can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}-groups or combinations thereof;

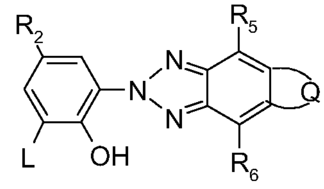
R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl or C_7-C_{15} phenylalkyl;

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; and

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22}.

Particularly preferred is a compound of formula (I) wherein

R_1 is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms; or

R_1 is a group  wherein L is alkylene of 1 to 12 carbon atoms, alkylidene of 2 to 12 carbon atoms, benzylidene, p-xylylene or cycloalkylene of 5 to 7 carbon atoms;

R_2 is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

R_2 is $-(CH_2)_m-CO-X_1-(Z)_p-Y-R_{15}$ wherein

X_1 is -O-,

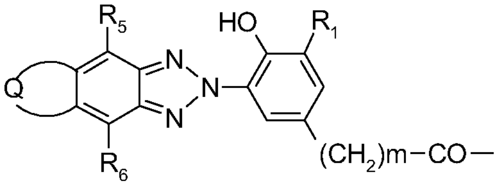
Y is -O- or a direct bond,

Z is C_2-C_{12}-alkylene, C_4-C_{12}alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or when Y is a direct bond, Z can additionally also be a direct bond;

m is 2,

p is 1,

R_{15} is hydrogen, C_1-C_{12}alkyl or a group



R_5 and R_6 are independently hydrogen or C_1-C_4alkyl.

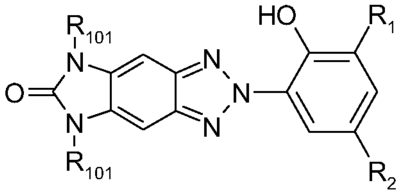
For instance, in the compound of formula (I)

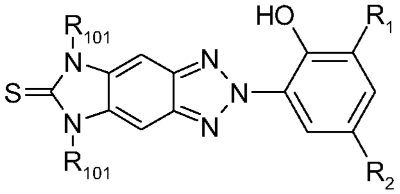
R_1 is hydrogen, straight or branched chain alkyl of 1 to 12 carbon atoms or phenylalkyl of 7 to 15 carbon atoms;

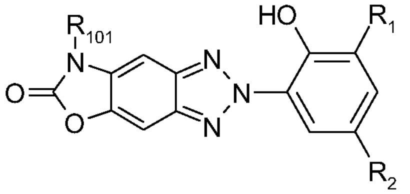
R_2 is straight or branched chain alkyl of 1 to 12 carbon atoms or phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms;

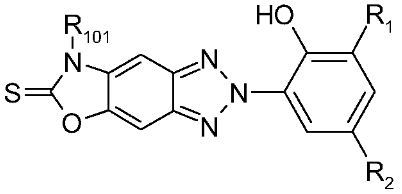
R_5 and **R_6** are hydrogen or C_1-C_4alkyl.

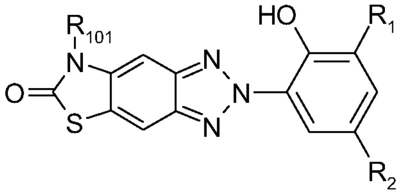
More preferred is a compound of formula (I) which is a compound according to formulae (a) to (i)

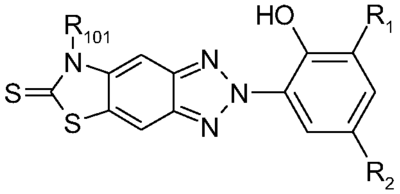

(a) ,

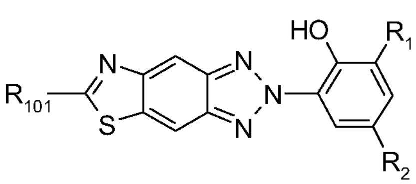

(b),

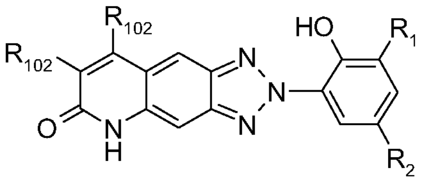

(c) ,

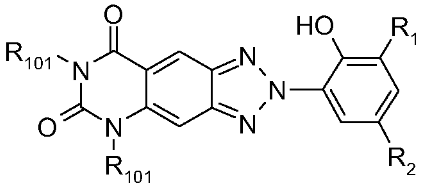

(d),

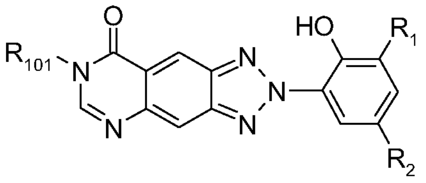

(e),

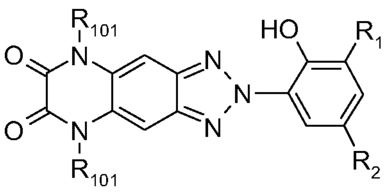

(f)


(g) ,


(h) and


(i) ,


or


wherein

R₁ is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms;

R₂ is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

R_2 is $-(CH_2)_m-CO-X_1-(Z)_p-Y-R_{15}$ wherein

X_1 is -O-,

Y is -O- or a direct bond,

Z is C_2-C_{12}-alkylene, C_4-C_{12}alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or when Y is a direct bond, Z can additionally also be a direct bond;

m is 2,

p is 1,

R_{15} is hydrogen, C_1-C_{12}alkyl;

R_5 and R_6 are hydrogen;

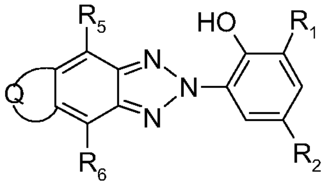
R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more –OH; or said straight or branched chain unsubstituted or substituted C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl or C_7-C_{15} phenylalkyl;

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; and

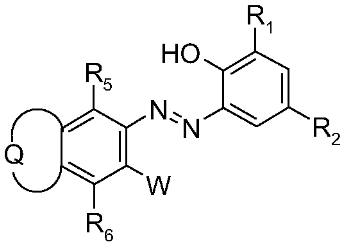
R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22}.

Another aspect of the invention is a process for the preparation of a compound of formula (I)



(I)

wherein the substituents R_1 to R_6 and Q are as defined above, which process comprises reacting a compound of formulae (III)

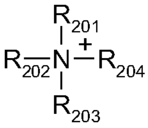


(III), wherein W is halogen or nitro

with an azide compound of formula (X)

$M^{n+}\,(\overset{-}{N_3})_r$ (X)

wherein

M is an n-valent metal cation,  $R_{202}-\overset{\displaystyle R_{201}}{\underset{\displaystyle R_{203}}{\overset{+}{N}}}-R_{204}$ or $P^+(R_{205})_4$,

R_{201}, **R_{202}**, **R_{203}** and **R_{204}** are each independently of the others hydrogen or C_1-C_{18}alkyl,
R_{205} is C_1-C_{18}alkyl, and
r is 1, 2 or 3.

Preferred reaction conditions of the process according to the invention are as follows:

The reaction can be carried out in the melt or in a solvent. Of special interest is a process for the preparation of compounds of formula I or II wherein the reaction is carried out in a solvent.

Suitable solvents are, for example, dipolar aprotic solvents, protic solvents, esters of aliphatic or aromatic carboxylic acids, ethers, halogenated hydrocarbons, aromatic solvents, amines and alkoxybenzenes.

Examples of dipolar aprotic solvents are dialkyl sulfoxides, for example dimethyl sulfoxide; carboxamides, for example formamide, dimethylformamide or N,N-dimethylacetamide; lactams, for example N-methylpyrrolidone; phosphoric amides, for example hexamethylphosphoric triamide; alkylated ureas, for example N,N'-dimethylethyleneurea, N,N'-dimethylpropyleneurea or N,N,N',N'-tetramethylurea; and nitriles, for example acetonitrile or benzonitrile.

Examples of protic solvents are polyalkylene glycols, for example polyethylene glycol; polyalkylene glycol monoethers, for example diethylene glycol monomethyl ether, and water, the latter on its own or in a single-phase or two-phase mixture with one or more of the solvents mentioned, it being possible also for phase transfer catalysts to be added, for example tetraalkylammonium salts, tetraalkylphosphonium salts or crown ethers. The same phase transfer catalysts can also be of use in solid/liquid form in the two-phase system.

Preferred esters of aliphatic or aromatic carboxylic acids are, for example, butyl acetate, cyclohexyl acetate and methyl benzoate.

Preferred ethers are, for example, dialkyl ethers, especially dibutyl ether, tetrahydrofuran, dioxane and (poly-)alkylene glycol dialkyl ethers.

Halogenated hydrocarbons are, for example, methylene chloride and chloroform.

Aromatic solvents are, for example, toluene, chlorobenzene and nitrobenzene.

Suitable amine solvents are, for example, triethylamine, tributylamine and benzyl-dimethylamine.

Preferred alkoxybenzenes are, for example, anisole and phenetole.

The process for the preparation of compounds of formula I can also be carried out in ionic or supercritical fluids, for example fluid carbon dioxide.

Of special interest is a process for the preparation of compounds of formula I wherein the reaction is carried out in a dipolar aprotic solvent.

The reaction temperatures can be varied within wide limits but are so selected that satisfactory conversion occurs, such temperatures preferably being from 10° to 200°C, especially from 20° to 150°C.

An analogous process for other benzotriazole compounds has already been disclosed in WO 02/24668.

Preference is given to a process for the preparation of compounds of formula I wherein the molar ratio of the amount of a compound of formula III to the amount of the azide compound of formula X is from 1 : 1 to 1 : 3, especially from 1 : 1 to 1 : 2, e.g. from 1 : 1 to 1 : 1.3. When functional side groups that are also able to react with azide are present, the excess of the azide compound of formula X is increased accordingly.

In a specific embodiment the reaction is carried out in the presence of a catalyst.

Such catalysts include, for example, copper(I) or copper(II) salts or other transition metal salts, based, for example, on iron, cobalt, nickel, palladium, platinum, gold or zinc. Instead of transition metal salts, the anions of which can be varied within wide limits, it is also possible to use metal complexes and metal complex salts of the same metals as catalysts. Preference is given to the use of copper(I) and copper(II) chlorides, bromides and iodides, and special preference to the use of copper(I) bromide.

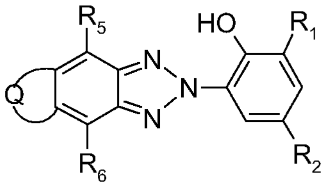
The catalyst is advantageously used in an amount of from 0.01 to 10 % by weight, especially from 0.1 to 5 % by weight, e.g. from 0.1 to 5 % by weight, based on the weight of the compound of formula III, IV or V employed.

The reaction can also be carried out in the presence of an additional base or in the presence of an alkaline pH buffer system. Suitable pH buffer systems include, for example, alkali metal or alkaline earth metal hydroxides; alkali metal or alkaline earth metal alcoholates; alkali metal or alkaline earth metal carboxylates, for example acetates or carbonates; alkali metal or alkaline earth metal phosphates; tertiary amines, for example triethylamine or tributylamine; and unsubstituted or substituted pyridines.

Some of the starting compounds of formula III are known from the literature or can be prepared analogously to the procedures described in the examples.

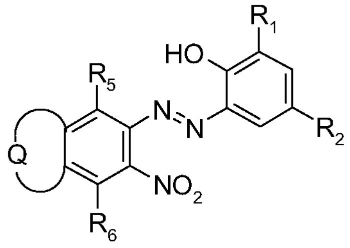
It is, however, also possible to prepare the instant benzotriazoles by conventional methods for preparing such compounds.

Consequently another aspect of the invention is an alternative process for the preparation of a compound of formula (I)



(I)

wherein the substituents R_1 to R_6 and Q are as above, which process comprises reacting a compound of formula (III)



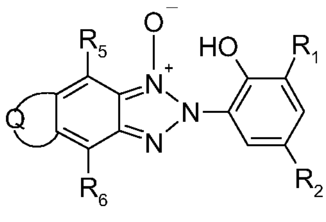
(III) under reductive conditions to a compound of formula (I).

The usual procedure involves the diazotization of a substituted o-nitroaniline followed by coupling the resultant diazonium salt with a substituted phenol and reduction of the azobenzene intermediate to the corresponding desired benzotriazole. Such processes are described, for example, in U.S. 5 276 161 and U.S. 5 977 219. The starting materials for these benzotriazoles are partly items of commerce or can be prepared by normal methods of organic synthesis.

Further methods for the preparation of benzotriazoles are for example given in Science of Synthesis 13.13, 575-576.

The reduction process can not only be carried out by hydration but also by other methods, such as for example described in EP 0 751 134. When a H-transfer is made, reagents, such as formic acid or its salts, phosphinic acid or its salts or an alkali metal or ammonium salt of hypophosphoric acid together with a catalyst may be useful. The catalyst is, for example, a precious metal.

A further aspect of the invention is a compound of formula (IV)



(IV)

wherein the substituents R_1 to R_6 and Q are as defined above.

These compounds are, when isolated, intermediates of the compounds according to formula (I).

The benzotriazoles of the present invention are generally useful as UV-absorbers in various substrates. Consequently a further aspect of the invention is a composition stabilized against light-induced degradation which comprises,
(a) an organic material subject to light-induced degradation, and
(b) a compound of formula I as described above.

In general the compound of formula I is present in an amount from 0.1% to 30%, preferably from 0.5% to 15% and more preferably from 1% to 10% by weight, based on the weight of the organic material.

In one aspect the organic material is a recording material.

The recording materials according to the invention are suitable for pressure-sensitive copying systems, photocopying systems using microcapsules, heat-sensitive copying systems, photographic materials and ink jet printing.

The recording materials according to the invention are distinguished by an unexpected improvement in quality, especially with regard to the fastness to light.

The recording materials according to the invention have the construction known for the particular use. They consist of a customary carrier, for example paper or plastic film, which has been coated with one or more layers. Depending on the type of the material, these layers contain the appropriate necessary components, in the case of photographic materials, for example, silver halide emulsions, dye couplers, dyes and the like. Material particularly suitable for ink jet printing has a layer particularly absorptive for ink on a customary carrier. Uncoated paper can also be employed for ink jet printing. In this case the paper acts at the same time as the carrier material and as the ink-absorbent layer. Suitable material for ink jet printing is, for example, described in U.S. Pat. No. 5,073,448 (incorporated herein by reference).

The recording material can also be transparent, as, for example, in the case of projection films.

The compounds of the formula I can be incorporated into the carder material as early as the production of the latter, in the production of paper, for example, by being added to the paper pulp. A second method of application is to spray the carder material with an aqueous solution of compounds of the formula I or to add the compounds to the coating composition.

Coating compositions intended for transparent recording materials suitable for projection cannot contain any particles which scatter light, such as pigments and fillers.

The dye-binding coating composition can contain a number of other additives, for example antioxidants, light stabilizers (including also UV absorbers which do not belong to the UV absorbers according to the invention), viscosity improvers, fluorescent brighteners, biocides and/or antistatic agents.

The coating composition is usually prepared as follows:
the water-soluble components, for example the binder, are dissolved in water and stirred together. The solid components, for example fillers and other additives already described, are dispersed in this aqueous medium. Dispersion is advantageously carded out by means of devices, for example ultrasonic samples, turbine stirrers, homogenizers, colloid mills, bead mills, sand mills, high-speed stirrers and the like. The compounds of the formula I can be incorporated easily into the coating composition.

The recording material according to this invention preferably contains 1 to 5000 mg/m^2, in particular 50-1200 mg/m^2, of a compound of the formula I.

As already mentioned, the recording materials according to the invention embrace a wide field. The compounds of the formula I can, for example, be employed in pressure-sensitive copying systems. They can be introduced either into the paper in order to protect the microencapsulated dye precursors there from light, or into the binder of the developer layer in order to protect the dyes formed there.

Photocopying systems using light-sensitive microcapsules which are developed by means of pressure are described in U.S. Pat. Nos. 4,416,966; 4,483,912; 4,352,200; 4,535,050; 4,5365,463; 4,551,407; 4,562,137 and 4,608,330, also in EP-A 139,479; EP-A 162,664; EP-

A 164,931; EP-A 237,024; EP-A 237,025 or EP-A 260,129. In all these systems the compounds can be put into the dye-receiving layer. The compounds can, however, also be put into the donor layer in order to protect the color formers from light.

Photographic materials which can be stabilized are photographic dyes and layers containing such dyes or precursors thereof, for example photographic paper and films. Suitable materials are, for example, described in U.S. Pat. No. 5,364,749 (incorporated therein by reference). The compounds of the formula I act here as a UV filter against electrostatic flashes. In color photographic materials couplers and dyes are also protected against photochemical decomposition.

The instant compounds can be used for all types of color photographic materials. For example, they can be employed for color paper, color reversal paper, direct-positive color material, color negative film, color positive film, color reversal film, etc. They are preferably used, inter alia, for photographic color material which contains a reversal substrate or forms positives.

Color-photographic recording materials usually contain, on a support, a blue-senitive and/or a green-sensitive and/or a red-sensitive silver-halide emulsion layer and, if desired, a protection layer, the compounds being, preferably, either in the green-sensitive or the red-sensitive layer or in a layer between the green-sensitive and the red-sensitive layer or in a layer on top of the silver-halide emulsion layers.

The compounds of the formula I can also be employed in recording materials based on the principles of photopolymerization, photoplasticization or the rupture of microcapsules, or in cases where heat-sensitive and light-sensitive diazonium salts, leuko dyes having an oxidizing agent or dye lactones having Lewis acids are used.

Furthermore, they can be employed in recording materials for dye diffusion transfer printing, thermal wax transfer printing and not matrix printing and for use with electrostatic, electrographic, electrophoretic, magnetographic and laser-electrophotographic printers and pen-plotters. Of the above, recording materials for dye diffusion transfer printing are preferred as, for example described in EP-A-507,734.

They can also be employed in inks, preferably for ink jet printing, as, for example, further described in U.S. Pat. No. 5,098,477 (incorporated herein by reference).

In another specific embodiment of the invention the organic material is a natural, semi-synthetic or synthetic polymer.

Examples of such polymers are given below.

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyvinylcyclohexane, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE).

Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different, and especially by the following, methods:

a) radical polymerisation (normally under high pressure and at elevated temperature).

b) catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either π- or σ-coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).

2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).

3. Copolymers of monoolefins and diolefins with each other or with other vinyl monomers, for example ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and mixtures thereof with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, ethylene/vinylcyclohexane copolymers, ethylene/cycloolefin copolymers (e.g. ethylene/norbornene like COC), ethylene/1-olefins copolymers, where the 1-olefin is generated in-situ; propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/vinylcyclohexene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of such copolymers with one another and with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinyl acetate copolymers (EVA), LDPE/ethylene-acrylic acid copolymers (EAA), LLDPE/EVA, LLDPE/EAA and alternating or random polyalkylene/carbon monoxide copolymers and mixtures thereof with other polymers, for example polyamides.

4. Hydrocarbon resins (for example C_5-C_9) including hydrogenated modifications thereof (e.g. tackifiers) and mixtures of polyalkylenes and starch.

Homopolymers and copolymers from 1.) - 4.) may have any stereostructure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereoblock polymers are also included.

5. Polystyrene, poly(p-methylstyrene), poly(α-methylstyrene).

6. Aromatic homopolymers and copolymers derived from vinyl aromatic monomers including styrene, α-methylstyrene, all isomers of vinyl toluene, especially p-vinyltoluene, all isomers of ethyl styrene, propyl styrene, vinyl biphenyl, vinyl naphthalene, and vinyl anthracene, and mixtures thereof. Homopolymers and copolymers may have any stereostructure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereoblock polymers are also included.

6a. Copolymers including aforementioned vinyl aromatic monomers and comonomers selected from ethylene, propylene, dienes, nitriles, acids, maleic anhydrides, maleimides, vinyl acetate and vinyl chloride or acrylic derivatives and mixtures thereof, for example styrene/butadiene, styrene/acrylonitrile, styrene/ethylene (interpolymers), styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength of styrene copolymers and another polymer, for example a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene such as styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.

6b. Hydrogenated aromatic polymers derived from hydrogenation of polymers mentioned under 6.), especially including polycyclohexylethylene (PCHE) prepared by hydrogenating atactic polystyrene, often referred to as polyvinylcyclohexane (PVCH).

6c. Hydrogenated aromatic polymers derived from hydrogenation of polymers mentioned under 6a.).

Homopolymers and copolymers may have any stereostructure including syndiotactic, isotactic, hemi-isotactic or atactic; where atactic polymers are preferred. Stereoblock polymers are also included.

7. Graft copolymers of vinyl aromatic monomers such as styrene or α-methylstyrene, for example styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene; styrene and maleic anhydride on

polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene and maleimide on polybutadiene; styrene and alkyl acrylates or methacrylates on polybutadiene; styrene and acrylonitrile on ethylene/propylene/diene terpolymers; styrene and acrylonitrile on polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 6), for example the copolymer mixtures known as ABS, MBS, ASA or AES polymers.

8. Halogen-containing polymers such as polychloroprene, chlorinated rubbers, chlorinated and brominated copolymer of isobutylene-isoprene (halobutyl rubber), chlorinated or sulfochlorinated polyethylene, copolymers of ethylene and chlorinated ethylene, epichlorohydrin homo- and copolymers, especially polymers of halogen-containing vinyl compounds, for example polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof such as vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.

9. Polymers derived from α,β-unsaturated acids and derivatives thereof such as polyacrylates and polymethacrylates; polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact-modified with butyl acrylate.

10. Copolymers of the monomers mentioned under 9) with each other or with other unsaturated monomers, for example acrylonitrile/ butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halide copolymers or acrylonitrile/ alkyl methacrylate/butadiene terpolymers.

11. Polymers derived from unsaturated alcohols and amines or the acyl derivatives or acetals thereof, for example polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; as well as their copolymers with olefins mentioned in 1) above.

12. Homopolymers and copolymers of cyclic ethers such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bisglycidyl ethers.

13. Polyacetals such as polyoxymethylene and those polyoxymethylenes which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.

14. Polyphenylene oxides and sulfides, and mixtures of polyphenylene oxides with styrene polymers or polyamides.

15. Polyurethanes derived from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as precursors thereof.

16. Polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, for example polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and with or without an elastomer as modifier, for example poly-2,4,4,-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, e.g. with polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and polyamides condensed during processing (RIM polyamide systems).

17. Polyureas, polyimides, polyamide-imides, polyetherimids, polyesterimids, polyhydantoins and polybenzimidazoles.

18. Polyesters derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, for example polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate, polyalkylene naphthalate (PAN) and polyhydroxybenzoates, as well as block copolyether esters derived from hydroxyl-terminated polyethers; and also polyesters modified with polycarbonates or MBS.

19. Polycarbonates and polyester carbonates.

20. Polyketones.

21. Polysulfones, polyether sulfones and polyether ketones.

22. Crosslinked polymers derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.

23. Drying and non-drying alkyd resins.

24. Unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low flammability.

25. Crosslinkable acrylic resins derived from substituted acrylates, for example epoxy acrylates, urethane acrylates or polyester acrylates.

26. Alkyd resins, polyester resins and acrylate resins crosslinked with melamine resins, urea resins, isocyanates, isocyanurates, polyisocyanates or epoxy resins.

27. Crosslinked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic or aromatic glycidyl compounds, e.g. products of diglycidyl ethers of bisphenol A and bisphenol F, which are crosslinked with customary hardeners such as anhydrides or amines, with or without accelerators.

28. Natural polymers such as cellulose, rubber, gelatin and chemically modified homologous derivatives thereof, for example cellulose acetates, cellulose propionates and cellulose butyrates, or the cellulose ethers such as methyl cellulose; as well as rosins and their derivatives.

29. Blends of the aforementioned polymers (polyblends), for example PP/EPDM, Polyamide/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR,

POM/acrylate, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS or PBT/PET/PC.

For example the polymer is a thermoplastic polymer.

In another embodiment the organic material is a coating, in particular an automotive coating.

Resins used in coatings are typically crosslinked polymers, for example, derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.

Also useful are unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low flammability.

Preferreably used are crosslinkable acrylic resins derived from substituted acrylates, for example epoxy acrylates, urethane acrylates or polyester acrylates.

Also possible are alkyd resins, polyester resins and acrylate resins crosslinked with melamine resins, urea resins, isocyanates, isocyanurates, polyisocyanates or epoxy resins.

Crosslinked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic or aromatic glycidyl compounds, e.g. products of diglycidyl ethers of bisphenol A and bisphenol F, which are crosslinked with customary hardeners such as anhydrides or amines, with or without accelerators.

The coating material may also be a radiation curable composition containing ethylenically unsaturated monomers or oligomers and a polyunsaturated aliphatic oligomer.

The alkyd resin lacquers which can be stabilized against the action of light in accordance with the instant invention are the conventional stoving lacquers which are used in particular for coating automobiles (automobile finishing lacquers), for example lacquers based on alkyd/melamine resins and alkyd/acrylic/melamine resins (see H. Wagner and H. F. Sarx, "Lackkunstharze" (1977), pages 99-123). Other crosslinking agents include glycouril resins, blocked isocyanates or epoxy resins.

It is also to be noted that the compounds of the present invention are applicable for use in non-acid catalyzed thermoset resins such as epoxy, epoxy-polyester, vinyl, alkyd, acrylic and polyester resins, optionally modified with silicon, isocyanates or isocyanurates. The epoxy and epoxy-polyester resins are crosslinked with conventional crosslinkers such as acids, acid anhydrides, amines and the like. Correspondingly, the epoxide may be utilized as the crosslinking agent for various acrylic or polyester resin systems that have been modified by the presence of reactive groups on the backbone structure.

When water-soluble, water miscible or water dispersible coatings are desired ammonium salts of acid groups present in the resin are formed. Powder coating composition can be prepared by reacting glycidyl methacrylate with selected alcohol components.

In a specific embodiment the above mentioned coating is applied over a substrate, which is sensitive to electromagnetic radiation of wavelengths greater than 380 nm.

A typical sensitive substrate is, for example, a cathodically deposited coating adhering to a metal substrate. Such coatings are typically used in the automotive industry.

Under sensitive to electromagnetic radiation of wavelengths greater than 380 nm there is understood UV or visible light, for example, in the wavelength range up to 440 nm, preferably up to 420 nm and in particular up to 410 nm.

For example the composition of the automotive coating comprises
a) a primer coat which is electrodeposited onto a metal substrate;
b) at least one pigmented base coat which is in direct contact with the primer coat, containing a compound of formula (I) according to claim 1;
c) a clear coat which is in direct contact with the pigmented base coat, containing a UV-absorber selected from the group consisting of the s-triazines, the oxanilides, the hydroxybenzophenones, benzoates, the α-cyanoacrylates and the benzotriazoles different from those of formulae (I) and
d) optionally the clear coat contains also a compound of formula (I).

In a specific embodiment the above composition contains also in the primer coat a compound of formula (I).

In the various organic materials, in which the compounds of formula (I) are useful as stabilizers against the deleterious influence of UV and/or visible light further stabilizers and additives may be also present.

Examples are subsequently given.

1. Antioxidants

1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(α-methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example, 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.

1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.

1.4. Tocopherols, for example α-tocopherol, β-tocopherol, γ-tocopherol, δ-tocopherol and mixtures thereof (vitamin E).

1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)-disulfide.

1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α-methylcyclohexyl)-phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α-methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-(α,α-dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methyl-phenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis(5-tert-butyl-4-hydroxy2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra-(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.

1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)malonate, didodecylmercaptoethyl-2,2-bis (3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.

1.10. Triazine compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.

1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.

1.13. Esters of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.14. Esters of β-(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis-(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane; 3,9-bis[2-{3-(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]-undecane.

1.15. Esters of β-(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.17. Amides of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Naugard®XL-1, supplied by Uniroyal).

1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylaminophenol, 4-octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- and dialkylated tert-butyldiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- and dialkylated tert-butyl/tert-octylphenothiazines, a mixture of mono- and dialkylated tert-octyl-phenothiazines, N-allylphenothiazine, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene.

2. UV absorbers and light stabilizers

2.1. 2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chloro-benzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis-(α,α-dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)-carbonylethyl]-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenylbenzotriazole, 2,2'-methylene-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300; $\left[R-CH_2CH_2-COO-CH_2CH_2\right]_2$, where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazol-2-ylphenyl, 2-[2'-hydroxy-3'-(α,α-dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)-phenyl]-benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-(α,α-dimethylbenzyl)-phenyl]benzotriazole.

2.2. 2-Hydroxybenzophenones, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.

2.3. Esters of substituted and unsubstituted benzoic acids, for example 4-tert-butyl-phenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butylbenzoyl)resorcinol, benzoyl resorcinol, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2.4. Acrylates, for example ethyl α-cyano-β,β-diphenylacrylate, isooctyl α-cyano-β,β-diphenylacrylate, methyl α-carbomethoxycinnamate, methyl α-cyano-β-methyl-p-methoxycinnamate, butyl α-cyano-β-methyl-p-methoxy-cinnamate, methyl α-carbomethoxy-p-methoxycinnamate, N-(β-carbomethoxy-β-cyanovinyl)-2-methylindoline, neopentyl tetra(α-cyano-β,β-diphenylacrylate.

2.5. Nickel compounds, for example nickel complexes of 2,2'-thio-bis[4-(1,1,3,3-tetramethylbutyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenylundecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.

2.6. Sterically hindered amines, for example bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl) n-butyl-3,5-di-tert-butyl-4-hydroxybenzylmalonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)nitrilotriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate, 1,1'-(1,2-ethanediyl)-bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearyloxy-2,2,6,6-tetramethylpiperidine, bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-

piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyrrolidine-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidine-2,5-dione, a mixture of 4-hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine, a condensate of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine, a condensate of 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); a condensate of 1,6-hexanediamine and 2,4,6-trichloro-1,3,5-triazine as well as N,N-dibutylamine and 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [192268-64-7]); N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimide, N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimide, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxo-spiro[4,5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro-[4,5]decane and epichlorohydrin, 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidyloxycarbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, a diester of 4-methoxymethylenemalonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-piperidyl)]siloxane, a reaction product of maleic acid anhydride-α-olefin copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine, 2,4-bis[N-(1-cyclohexyloxy-2,2,6,6-tetramethylpiperidine-4-yl)-N-butylamino]-6-(2-hydroxyethyl)amino-1,3,5-triazine, 1-(2-hydroxy-2-methylpropoxy)-4-octadecanoyloxy-2,2,6,6-tetramethylpiperidine, 5-(2-ethylhexanoyl)-oxymethyl-3,3,5-trimethyl-2-morpholinone, Sanduvor (Clariant; CAS Reg. No. 106917-31-1], 5-(2-ethylhexanoyl)oxymethyl-3,3,5-trimethyl-2-morpholinone, the reaction product of 2,4-bis-[(1-cyclohexyloxy-2,2,6,6-piperidine-4-yl)butylamino]-6-chloro-s-triazine with N,N’-bis(3-aminopropyl)ethylenediamine), 1,3,5-tris(N-cyclohexyl-N-(2,2,6,6-tetramethylpiperazine-3-one-4-yl)amino)-s-triazine, 1,3,5-tris(N-cyclohexyl-N-(1,2,2,6,6-pentamethylpiperazine-3-one-4-yl)-amino)-s-triazine.

<u>2.7. Oxamides</u>, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture

with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

2.8. 2-(2-Hydroxyphenyl)-1,3,5-triazines, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxyphenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(4-[2-ethylhexyloxy]-2-hydroxyphenyl)-6-(4-methoxyphenyl)-1,3,5-triazine.

3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl)hydrazine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

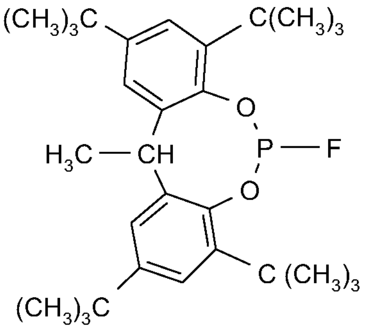
4. Phosphites and phosphonites, for example triphenyl phosphite, diphenylalkyl phosphites, phenyldialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearylpentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, bis(2,4-dicumylphenyl)pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)-pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-

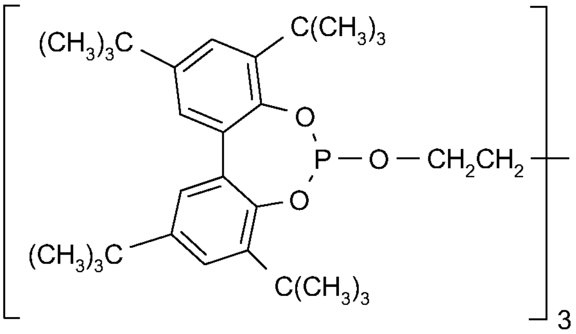
isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl)methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl)ethyl phosphite, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, 2,2',2''-nitrilo-[triethyltris(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite, 5-butyl-5-ethyl-2-(2,4,6-tri-tert-butylphenoxy)-1,3,2-dioxaphosphirane.

The following phosphites are especially preferred:

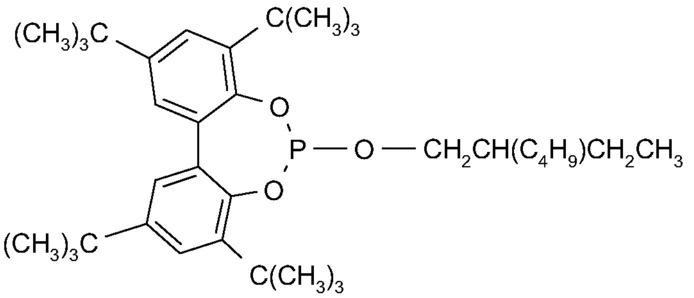
Tris(2,4-di-tert-butylphenyl) phosphite (Irgafos®168, Ciba Specialty Chemicals Inc.), tris(nonylphenyl) phosphite,

(A)

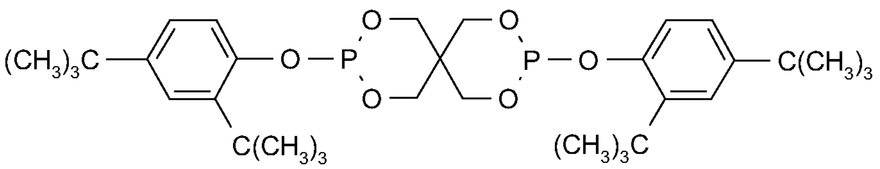




(B)



(C)



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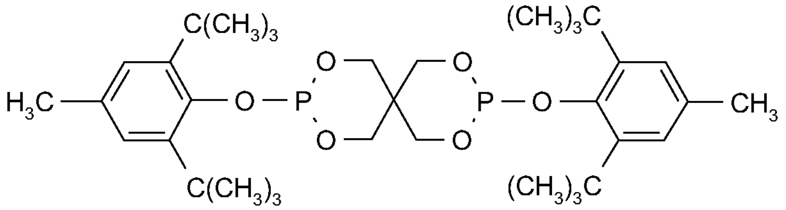
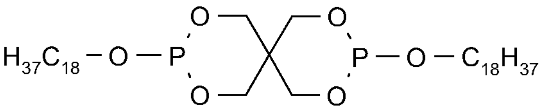
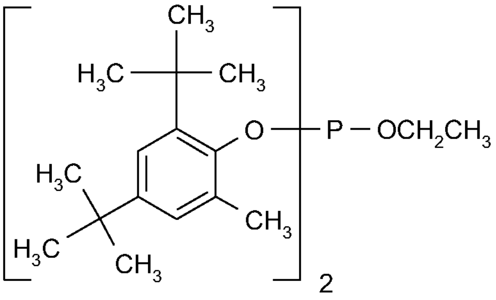


(E)

(F)





(G)

5. Hydroxylamines, for example N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-dilaurylhydroxylamine, N,N-ditetradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N-heptadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

6. Nitrones, for example, N-benzyl-alpha-phenylnitrone, N-ethyl-alpha-methylnitrone, N-octyl-alpha-heptylnitrone, N-lauryl-alpha-undecylnitrone, N-tetradecyl-alpha-tridecylnnitrone, N-hexadecyl-alpha-pentadecylnitrone, N-octadecyl-alpha-heptadecylnitrone, N-hexadecyl-alpha-heptadecylnitrone, N-ocatadecyl-alpha-pentadecylnitrone, N-heptadecyl-alpha-heptadecylnitrone, N-octadecyl-alpha-hexadecylnitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

7. Thiosynergists, for example dilauryl thiodipropionate, dimistryl thiodipropionate, distearyl thiodipropionate or distearyl disulfide.

8. Peroxide scavengers, for example esters of β-thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercapto-

benzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis(β-dodecylmercapto)propionate.

9. Polyamide stabilizers, for example copper salts in combination with iodides and/or phosphorus compounds and salts of divalent manganese.

10. Basic co-stabilizers, for example melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids, for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zinc pyrocatecholate.

11. Nucleating agents, for example inorganic substances, such as talcum, metal oxides, such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds, such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds, such as ionic copolymers (ionomers). Especially preferred are 1,3:2,4-bis(3',4'-dimethylbenzylidene)sorbitol, 1,3:2,4-di(paramethyldibenzylidene)sorbitol, and 1,3:2,4-di(benzylidene)sorbitol.

12. Fillers and reinforcing agents, for example calcium carbonate, silicates, glass fibres, glass beads, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibers.

13. Other additives, for example plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flameproofing agents, antistatic agents and blowing agents.

14. Benzofuranones and indolinones, for example those disclosed in U.S. 4,325,863; U.S. 4,338,244; U.S. 5,175,312; U.S. 5,216,052; U.S. 5,252,643; DE-A-4316611; DE-A-4316622; DE-A-4316876; EP-A-0589839, EP-A-0591102; EP-A-1291384 or 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butylbenzofuran-2-one, 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one, 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one], 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one, 3-(4-acetoxy-3,5-di-

methylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,4-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(2,3-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(2-acetyl-5-isooctylphenyl)-5-isooctylbenzofuran-2-one.

Preferably the compositions above contain additionally a sterically hindered amine stabilizer and/or a UV absorber selected from the group consisting of the s-triazines, the oxanilides, the hydroxybenzophenones, benzoates, the α-cyanoacrylates and the benzotriazoles different from those of formulae (I) as described above.

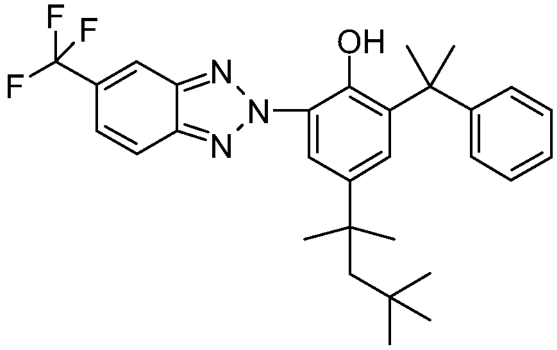
When additional UV-absorbers are added they are preferably added in an amount from 0.1% to 30%, more preferably from 0.5% to 15% and most preferably from 1% to 10% by weight, based on the weight of the organic material.

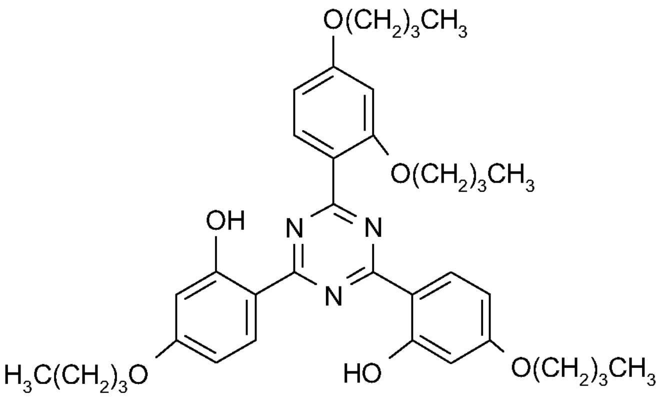
When a hindered amine light stabilizer is additionally added it is preferably added in an amount from 0.1% to 10%, more preferably from 0.5% to 5% and most preferably from 1% to 3% by weight, based on the weight of the organic material.

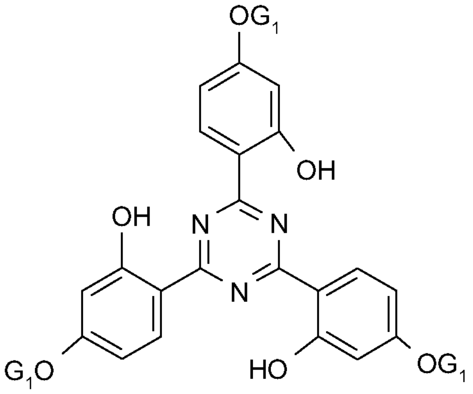
The total amount of UV-absorber of formula I and other UV-absorbers and/or hindered amine stabilizer is for example from 0.5% to 15% by weight, based on the weight of the organic material.

Examples for the hindered amine light stabilizers and UV-absorbers of the different classes are given above.

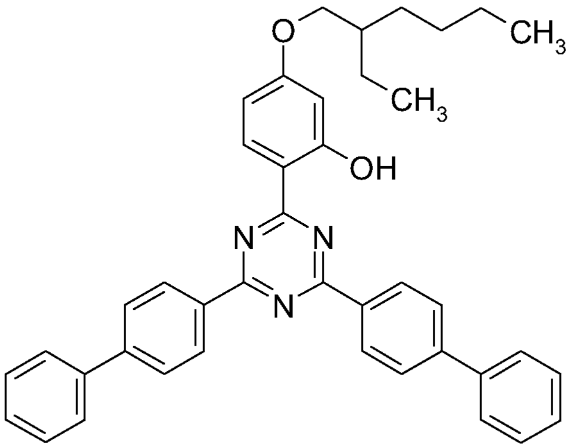
Particularly preferred UV-absorbers are the following s-triazines and benzotriazoles:

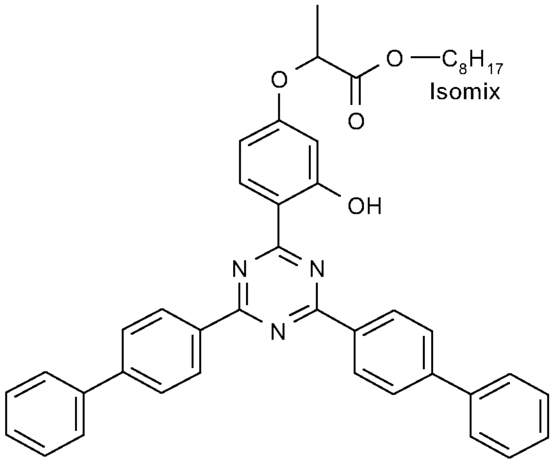


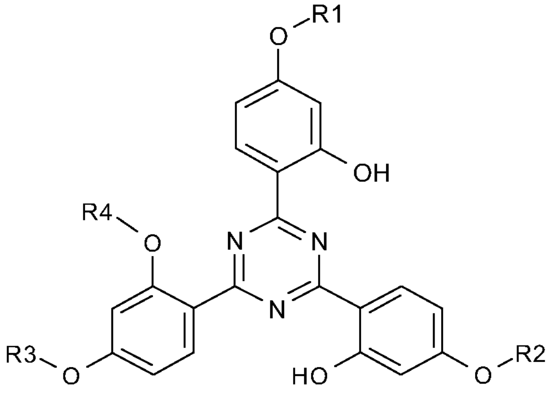




; G1= $CH(CH_3)\text{-}COO\text{-}C_2H_5$





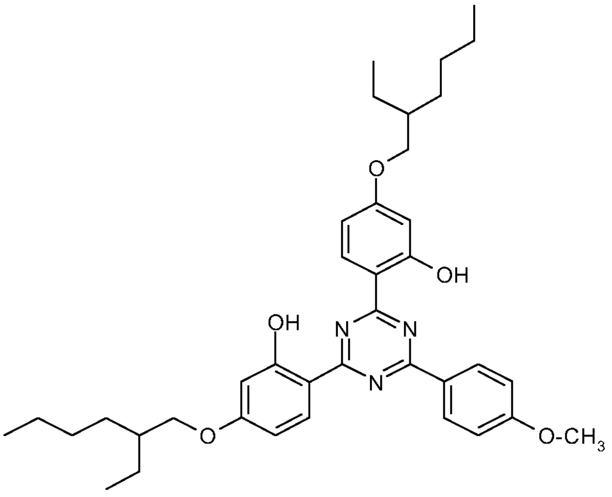


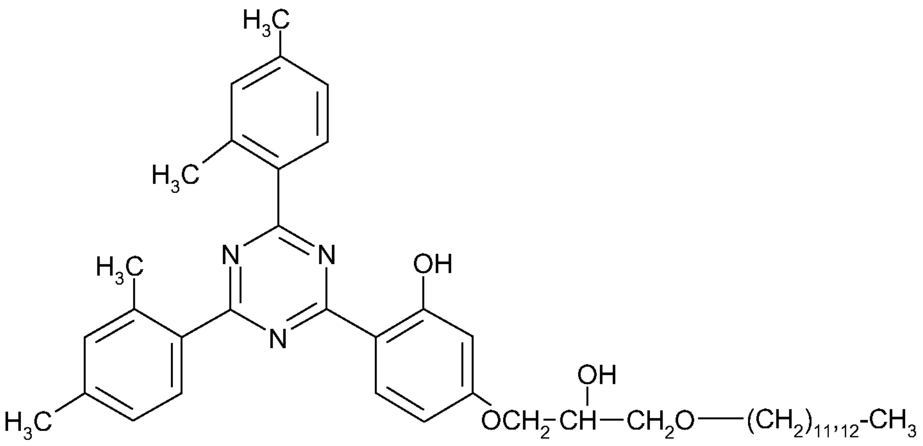
= a mixture of

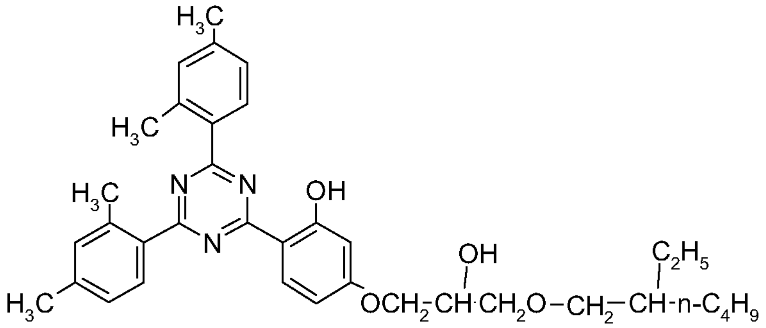
a) R1 = R2 = $CH(CH_3)$-COO-C_8H_{17}, R3= R4 =H;

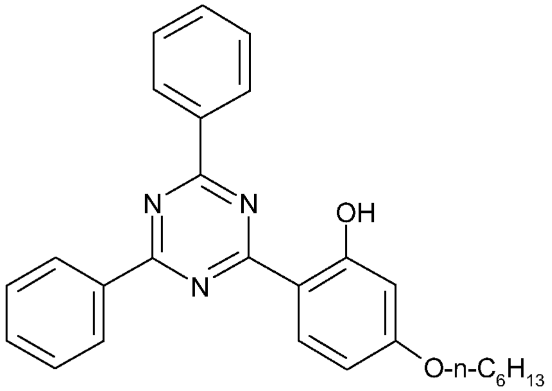
b) R1 = R2 = R3 = $CH(CH_3)$-COO-C_8H_{17}, R4 =H;

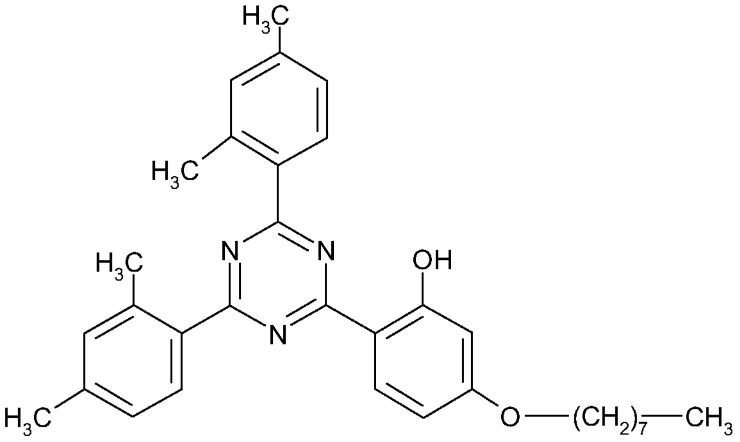
c) R1 = R2 = R3 = R4 = $CH(CH_3)$-COO-C_8H_{17}

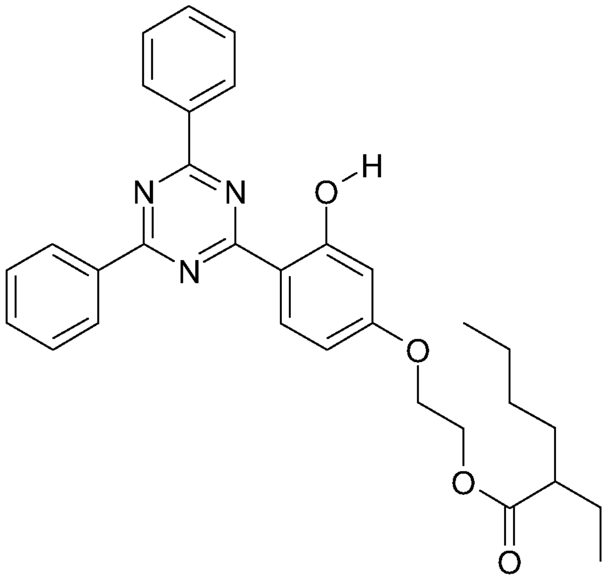



H3C
H3C
H3C
N
N
N
OH
H3C
OH
OCH2-CH—CH2O——(CH2)11,12-CH3


H3C
H3C
H3C
N
N
N
OH
H3C
OH
C2H5
OCH2-CH·CH2O-CH2-CH-n-C4H9


N
N
N
OH
O-n-C6H13





The hydroxyphenyl triazine UV-absorbers are known and are partially items of commerce.

The most suitable benzotriazole UV-absorbers are commercially available under the Trade Names TINUVIN 384 (RTM), TINUVIN 928 (RTM), TINUVIN 900 (RTM), TINUVIN 328 (RTM) and TINUVIN 1130 (RTM).

The sterically hindered amine compounds of component (c) are preferably selected from the group consisting of the following commercial products: DASTIB 845 (RTM), TINUVIN 770 (RTM), TINUVIN 765 (RTM), TINUVIN 144 (RTM), TINUVIN 123 (RTM), TINUVIN 111 (RTM), TINUVIN 783 (RTM), TINUVIN 791 (RTM), TINUVIN 123 (RTM), TINUVIN 292 (RTM), TINUVIN 152 (RTM), TINUVIN 144 (RTM), MARK LA 52 (RTM), MARK LA 57 (RTM), MARK LA 62

(RTM), MARK LA 67 (RTM), HOSTAVIN N 20 (RTM), HOSTAVIN N 24 (RTM), SANDUVOR 3050 (RTM), SANDUVOR 3058 (RTM), DIACETAM 5 (RTM), SUMISORB TM 61 (RTM), UVINUL 4049 (RTM), SANDUVOR PR 31(RTM), GOODRITE UV 3034 (RTM), GOODRITE UV 3150 (RTM), GOODRITE UV 3159 (RTM), GOODRITE 3110 x 128 (RTM), UVINUL 4050 H (RTM), CHIMASSORB 944 (RTM), CHIMASSORB 2020 (RTM), CYASORB UV 3346 (RTM), CYASORB UV 3529 (RTM), DASTIB 1082 (RTM), CHIMASSORB 119 (RTM), UVASIL 299 (RTM), UVASIL 125 (RTM), UVASIL 2000 (RTM), UVINUL 5050 H (RTM), LICHTSCHUTZSTOFF UV 31 (RTM), LUCHEM HA B 18 (RTM), MARK LA 63 (RTM), MARK LA 68 (RTM), UVASORB HA 88 (RTM), TINUVIN 622 (RTM), HOSTAVIN N 30 (RTM) and FERRO AM 806 (RTM).

Particularly preferred are TINUVIN 770 (RTM), TINUVIN 292 (RTM), TINUVIN 123 (RTM), TINUVIN 144 (RTM) and TINUVIN 152 (RTM).

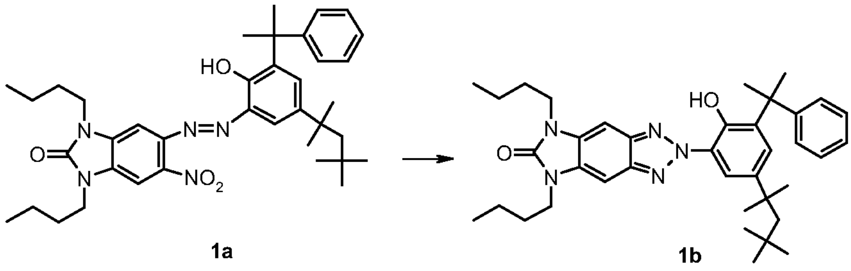
Yet another aspect of the invention is the use of a compound of formula I as ultraviolet (UV) and visible (VIS) light absorber in organic materials.

The definitions and preferences given for the compounds apply also for the other aspects of the invention.

The following examples illustrate the invention.

A) Preparation examples

Example A1. Preparation of compound **1b**



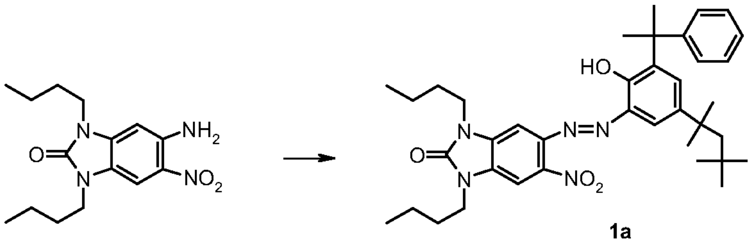
A stirred mixture of compound **1a** (40.0g, 62mmol), sodium azide (99%; 6.1g, 93mmol) and 1-methyl-2-pyrrolidinone (150ml) is heated to 160°C. The temperature is maintained and the progress of reaction is monitored by TLC (6.5 hours). The dark solution is cooled over night to room temperature followed by the addition of water (1000ml) and ethyl acetate (1500ml). The water phase is split off and washed with ethyl acetate(1x250ml). The combined organic phases are washed with water (3x100ml), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is dissolved in hot methanol. After cooling the slightly pink precipitate is filtered off and recrystallized twice from acetone.

Yield 32.8g (54mmol, 57%)

Melting point: 148°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 364 (33520)

The above mentioned intermediate **1a** is prepared as follows:

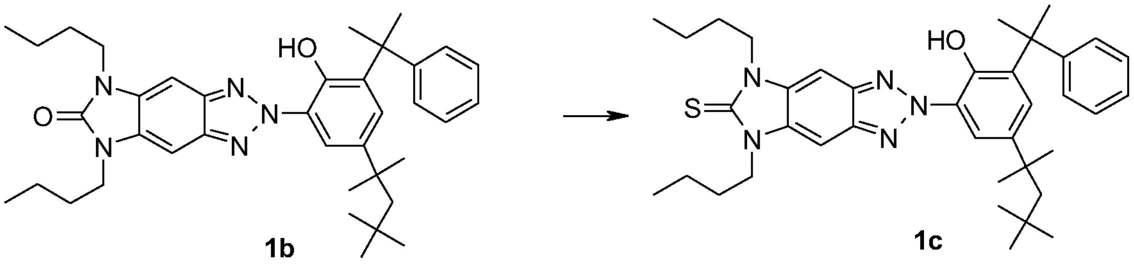


1a

Sodium nitrite (4 molar in water; 32.5ml, 130mmol) is slowly added between 0°C and 5°C to a stirred solution of 5-amino-1,3-dibutyl-6-nitro-benzimidazol-2-one (99.5%, 40.0g, 130mmol) in acetic acid (300ml) containing hydrochloric acid (32% in water; 35g). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (one hour), stirring is continued for 1.5 hours. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 46.2g, 135.0mmol) in methanol (250ml) containing sodium hydroxide microprills (5.5g, 137.5mmol). During the addition, the temperature of the reaction mixture is kept between -20°C and -6°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water; total of 350ml). After the addition is complete (2 hours), the cooling bath is removed and the red suspension stirred overnight. After adding water (50ml) and toluene (200ml) the pH is brought to less than seven using hydrochloric acid (30% in water). The water phase is split off and washed with toluene (2x100ml). The combined organic phases are washed with water, dried (Na_2SO_4), filtered and the solvent evaporated. Methanol (200ml) is added and the resulting solution cooled by means of an ice bath. Compound **1a** crystallizes as red solid, which is filtered off, washed with cold methanol and dried. Yield 63.9g (99mmol, 77%).

Melting point: 156°C

Example A2. Preparation of compound **1c**



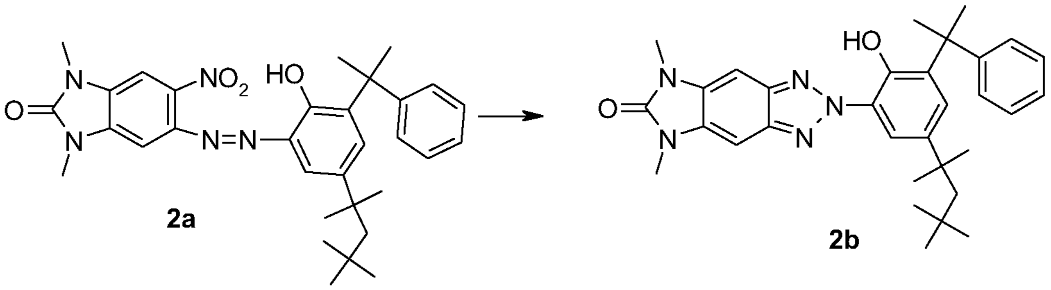
A stirred mixture of compound **1b** (10.0g, 16.4mmol), Lawesson Reagent (98%; 16.2g, 39.3mmol) and xylene (500ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled to room temperature followed by the addition of water and ethyl acetate. The water phase is split off and washed with ethyl acetate. The combined organic phases are washed with water (3x), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 2:1) and the obtained product is recrystallized from isopropanol.

Yield 7.6g (12.1mmol, 74%)

Melting point: 137°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 388 (56795)

Example 3. Preparation of compound **2b**

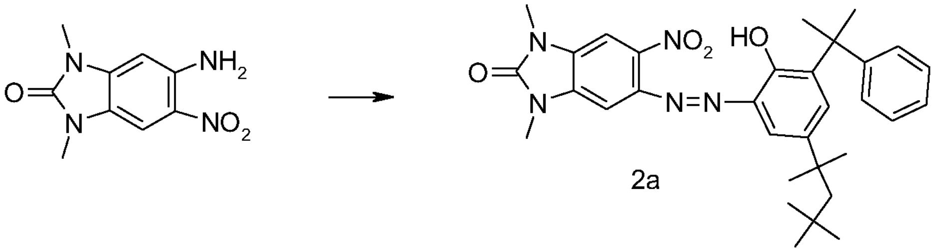


A stirred mixture of compound **2a** (8.9g; 15.8mmol), sodium azide (99%; 1.5g; 23.1mmol) and 1-methyl-2-pyrrolidinone (160ml) is heated to 160°C. The temperature is maintained and the progress of reaction is monitored by TLC (16 hours). The dark solution is added in a stirred mixture of ice and water. The precipitate is filtered off and dried.
Yield 5.7g (10.8mmol, 69%)

Melting point: 187°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 362 (35288)

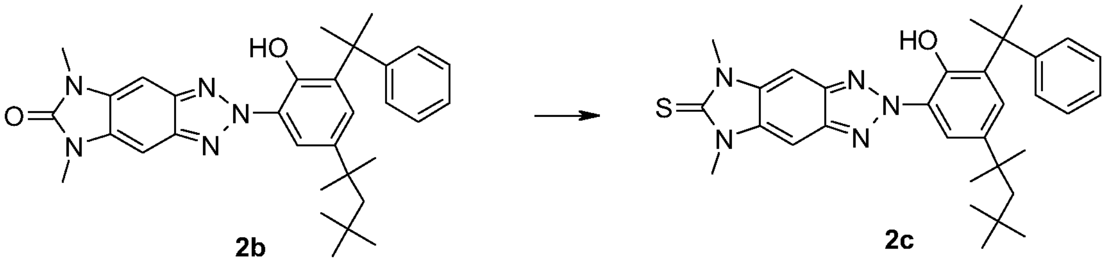
The above mentioned intermediate **2a** is prepared as follows:



Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of 5-amino-1,3-dimethyl-6-nitro-benzimidazol-2-one (8g, 36mmol) in acetic acid (80ml) containing hydrochloric acid (32% in water; 10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (15 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -30°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 11.6g, 35.7mmol) in methanol (50ml) containing sodium hydroxide microprills (1.4g, 36mmol). During the addition, the temperature of the reaction mixture is kept between -20°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water; total of 130ml). After the addition is complete (1 hour), the cooling bath is removed and the red suspension stirred overnight. After adding brine (200ml) and ethyl acetate (300ml), the water phase is split off and washed with ethyl acetate (1x50ml). The combined organic phases are washed with water, dried (Na_2SO_4), filtered and the solvent evaporated. Methanol (200ml) is added and the resulting solution cooled by means of an ice bath. Compound **2a** crystallizes as a brownish red solid, which is filtered off, washed with cold methanol and dried. Yield 8.3g (14.9mmol, 41%).

Melting point: 269°C

Example 4. Preparation of compound 2**c**

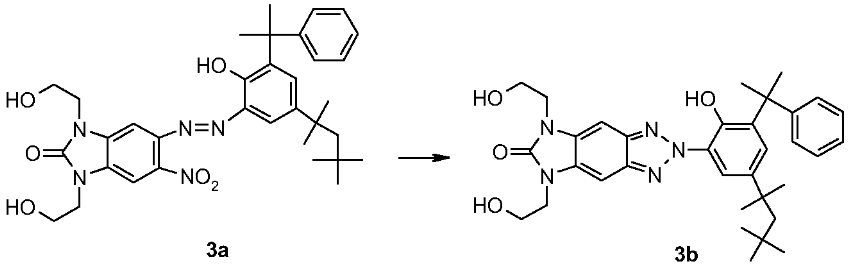


A stirred mixture of compound 2**b** (2.7g, 5.1mmol), Lawesson Reagent (98%; 5.2g, 12.5mmol) and xylene (100ml) is heated to 135°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is concentrated to the half, cooled and filtered. The filtrate is concentrated to dryness and the residue is chromatographed on a silica gel column (hexane / ethyl acetate 1:1) and the obtained product is recrystallized from isopropanol.
Yield 2.4g (4.2mmol, 83%)

Melting point: 196°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 386 (49570)

Example 5. Preparation of compound **3b**



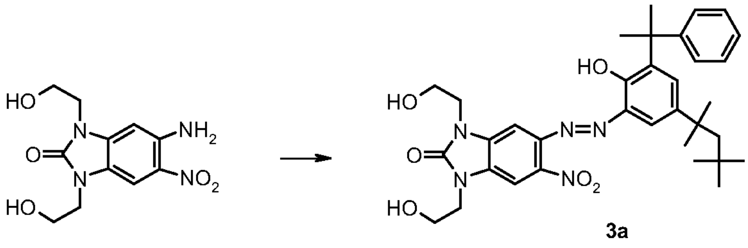
A stirred mixture of compound **3a** (5.8g; 9.4mmol), sodium azide (99%; 0.91g; 14.1mmol) and 1-methyl-2-pyrrolidinone (150ml) is heated to 160°C. The temperature is maintained and the progress of reaction is monitored by TLC (16hours). The dark solution is added to a stirred mixture of ice and water. The precipitate is filtered off and chromatographed on a silica gel column (ethyl acetate / methanol 98:2)

Yield 2.3g (39.3mmol, 42%)

Melting point: 197°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 363 (33922)

The above mentioned intermediate **3a** is prepared as follows:

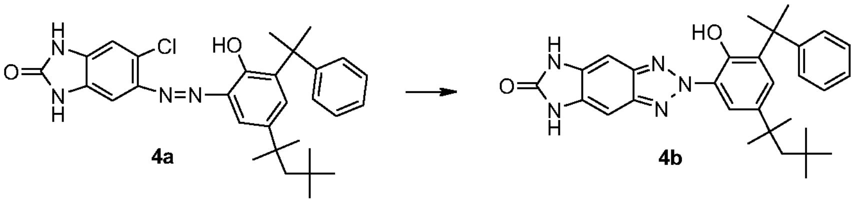


Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of 5-amino-1,3-dihydroxyethyl-6-nitro-benzimidazol-2-one (8.9g, 3.5mmol) in acetic acid (70ml) containing hydrochloric acid (32% in water; 10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (15 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -20°C to a stirred

solution of 2-cumyl-4-t-octylphenol (95%; 10.7g, 31.5mmol) in methanol (50ml) containing sodium hydroxide microprills (1.3g, 31.5mmol). During the addition, the temperature of the reaction mixture is kept between -20°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water; total of 100ml). After the addition is complete (1.5 hours), the cooling bath is removed and the red suspension stirred overnight. After adding brine (200ml) the red residue is filtered off and chromatographed on a silica gel column (hexane / ethyl acetate 1:9) and the obtained product is recrystallized from methanol.
Yield 7.1g (11.5mmol, 37%)

Melting point: 211°C

Example 6: Preparation of compound 4**b**

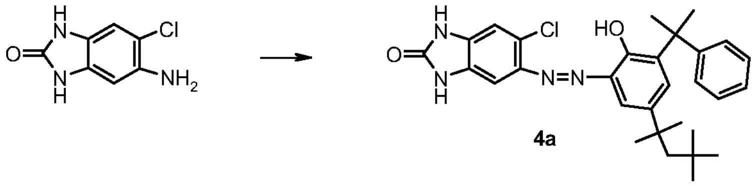


A stirred mixture of compound **4a** (10.4g; 20mmol), sodium azide (99%; 1.7g; 26mmol), copper(I) bromide (0.29g; 2mmol) and dimethyl formamide (40ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (3.5 hours). The dark solution is added to a stirred mixture of ice and water. The precipitate is filtered off and chromatographed on a silica gel column (ethyl acetate / methanol 12:1)
Yield 2.5g (5mmol, 25%)

^{1}H-NMR (400MHz, CDCl$_3$), δ (ppm): 11.23 (s broad, 1H), 9.01 (s broad, 2H), 8.26 (s, 1H), 7.61 (s, 1H), 7.33 (s, 2H), 7.31 - 7.29 (m, 4H), 7.20 - 7.17 (m, 1H), 1.82 (s, 6H), 1.79 (s, 2H), 1.28 (s, 6H), 0.80 (s, 9H).

UV-vis (dioxane), λ_{max} / nm (ε / dm^3 mol^{-1} cm^{-1}): 359 (35801)

The above mentioned intermediate **4a** is prepared as follows:



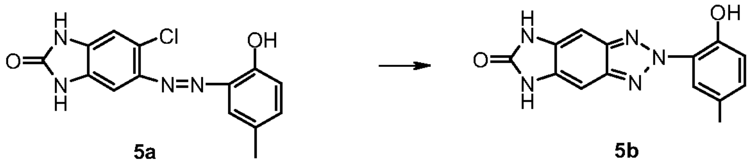
4a

Sodium nitrite (4 molar in water; 25ml, 100mmol) is slowly added between 0°C and 5°C to a stirred suspension of 5-amino-6-chloro-benzimidazol-2-one (18.36g, 100mmol) in acetic acid (250ml) containing hydrochloric acid (32% in water; 40ml) and water (80ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (0.5 hours), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 34.15, 100mmol) in methanol / xylene (85:15; 200ml) containing sodium hydroxide microprills (4g, 100mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. The red suspension is filtered off and the residue solved in hot isopropanol. After cooling, the liquid is filtered an evaporated to dryness. The solid is dissolved in less hot ethyl acetate, precipitated with water.

Yield 33.1g (63.8mmol, 64%)

^{1}H-NMR (400MHz, $CDCl_3$), δ (ppm): 13.02 (s broad, 1H), 9.51 (s broad, 1H), 9.48 (s broad, 1H), 7.77 (d, 1H), 7.64 (d like, 1H), 7.57 (d, 1H), 7.31 - 7.25 (m, 4H), 7.18 - 7.14 (m, 1H), 7.13 (d like, 1H), 1.86 (s, 2H), 1.83 (s, 6H), 1.51 (s, 6H), 0.84 (s, 9H).

Example 7 : Preparation of compound 5**b**



5a 5b

A stirred mixture of compound **5a** (96%; 20.0g; 66mmol), sodium azide (99%; 5.65g; 85.8mmol), copper(I) bromide (495mg; 0.7mmol) and 1-methyl-2-pyrrolidinone (75ml) is heated to 150°C. The temperature is maintained and the progress of reaction is monitored by TLC (2 days). The dark solution is added to a stirred mixture of ice and water. The precipitate is filtered off and the filter cake is washed in a soxhlet apparatus with 600 ml of ethyl acetate for 3 days. Then the product is extracted from the thimble by another 3 days of soxhlet extraction with 500 ml dioxane. The dioxane extract is concentrated to the half, the product is precipitated by addition of water and dried.

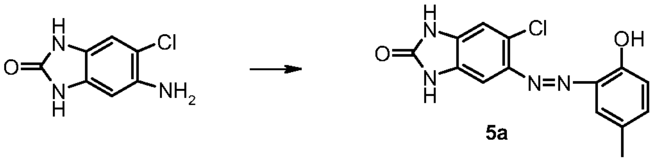
Yield: 2.5g (8.9 mmol, 13%)

Melting point: 300°C decomposition

^{1}H-NMR (300MHz, DMSO-d$_6$), δ (ppm): 10.02 (s broad, 2H), 10.63 (s broad, 1H), 7.76 – 7.75 (d, 1H), 7.28 (s, 2H) 7.17-7.15 (dd, 1H), 7.04-7.02 (d, 1H), 2.31 (s, 3H).

UV-vis (dioxane), λ_{max} / nm (ε / dm^3 mol^{-1} cm^{-1}): 355 (37735)

The above mentioned intermediate **5a** is prepared as follows:

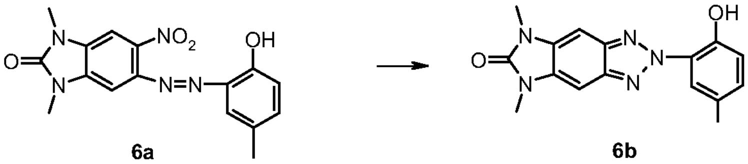


Sodium nitrite (4 molar in water; 20ml, 80mmol) is slowly added between 0°C and 5°C to a stirred suspension of 5-amino-6-chloro-benzimidazol-2-one (13.77g, 75mmol) in acetic acid (200ml) containing hydrochloric acid (32% in water; 20ml) and water (40ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (0.5 hours), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of p-cresol (99%; 8.19g, 75mmol) in methanol / xylene (85:15; 125ml) containing sodium hydroxide microprills (3g, 75mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the thick suspension stirred overnight. The suspension is filtered off and dryed.

Yield 17.1g (56.5mmol, 73.3%; HPLC-MS: MW 301.9 / 95.6% Area UV)

Melting point: 290°C decomposition

Example 8: Preparation of compound **6b**



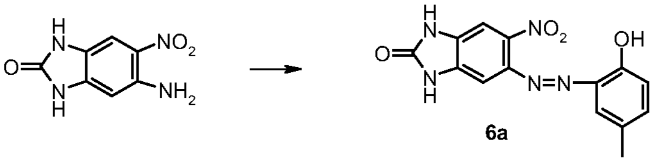
A stirred mixture of compound 6**a** (3.4g; 10mmol), sodium azide (99%; 0.85g; 13mmol) and 1-methyl-2-pyrrolidinone (25ml) is heated to 160°C. The temperature is maintained and the progress of reaction is monitored by TLC (16hours). The dark solution is added to a stirred mixture of ice and water. The residue is filtered off and the product is extracted from the residue by Soxhlet extraction with 400 ml ethyl acetate. After evaporation of the ethyl acetate the residue is suspended in 30% aqueous sodium hydroxide (50ml) and ethanol(50ml) and heated to reflux while stirring. After cooling to room temperature a 32% aqueous hydrogen chloride solution is added (30ml) and the precipitate is filtered of. The procedure is repeated by refluxing the obtained residue in 30% aqueous sodium hydroxide (30ml), ethanol(30ml) and water (10ml) and finally adding 32% aqueous hydrogen chloride solution (30ml). The precipitate is filtered off, washed with water and dried.

Yield 0.82g (26.5mmol, 27%).

Melting point: 280°C

UV-vis (dioxane), λ_{max} / nm (ε / dm^{3}mol^{-1}cm^{-1}): 357 (38187)

The above mentioned intermediate **6a** is prepared as follows:

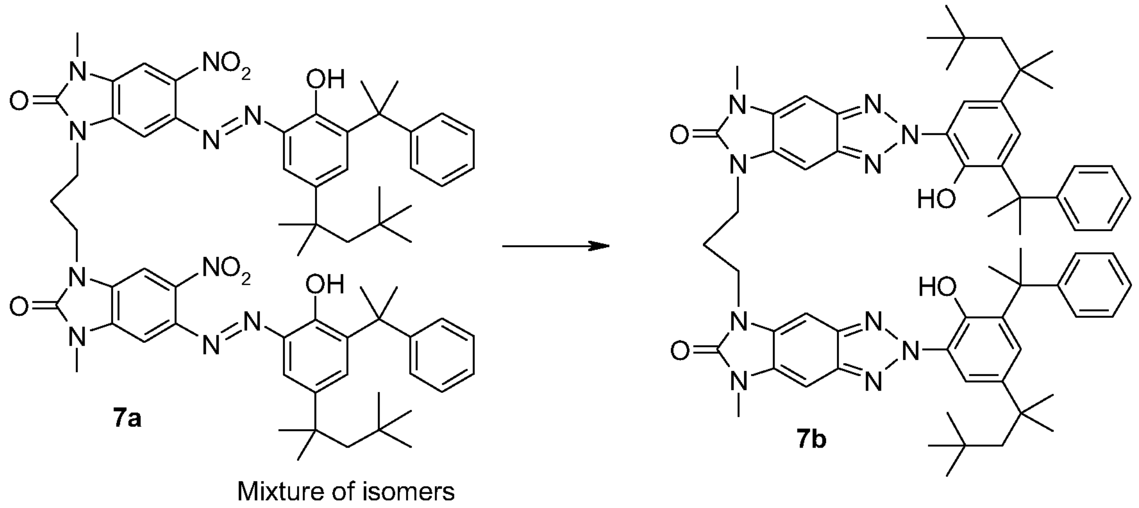


Sodium nitrite (4 molar in water; 27.5ml, 110mmol) is slowly added between 0°C and 5°C to a stirred suspension of 5-amino-1,3-dimethyl-6-nitro-benzimidazol-2-one (22.2g, 100mmol) in acetic acid (250ml) containing hydrochloric acid (32% in water; 25ml) and water (50ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (45 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of p-cresol (99%; 10.95g, 100mmol) in methanol (170ml) containing sodium hydroxide microprills (4g, 100mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water; 350ml). After the addition is complete (1 hour; end pH 6.3), the cooling bath is removed and the orange suspension stirred overnight. The suspension is filtered off, washed with water and dryed.

Yield 31.4g (95mmol, 95%)

Melting point: 281°C

Example 9. Preparation of compound **7b** CG45-0085 (FRK258/4)



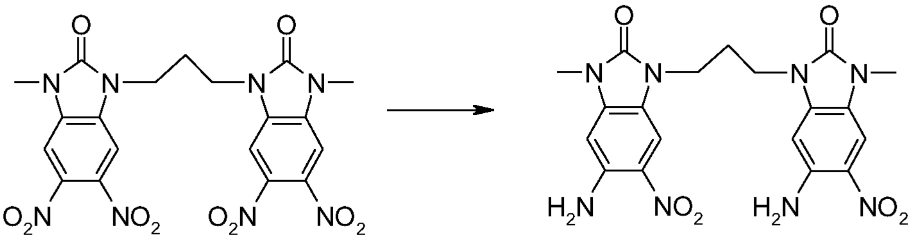
A stirred mixture of crude compound **7a** (20.0g) prepared as described below, sodium azide (99%; 3.6g, 55mmol) and 1-methyl-2-pyrrolidinone (75ml) is heated to 180°C. The temperature is maintained and the progress of reaction is monitored by TLC (6 hours). The dark solution is cooled over night to room temperature followed by the addition of water (100ml) and toluene (300ml).The organic phase is washed with water (1x50ml), and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 1:1).

Yield 5.0g (4.7mmol, 26%)

Melting point: 263°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 363 (66017)

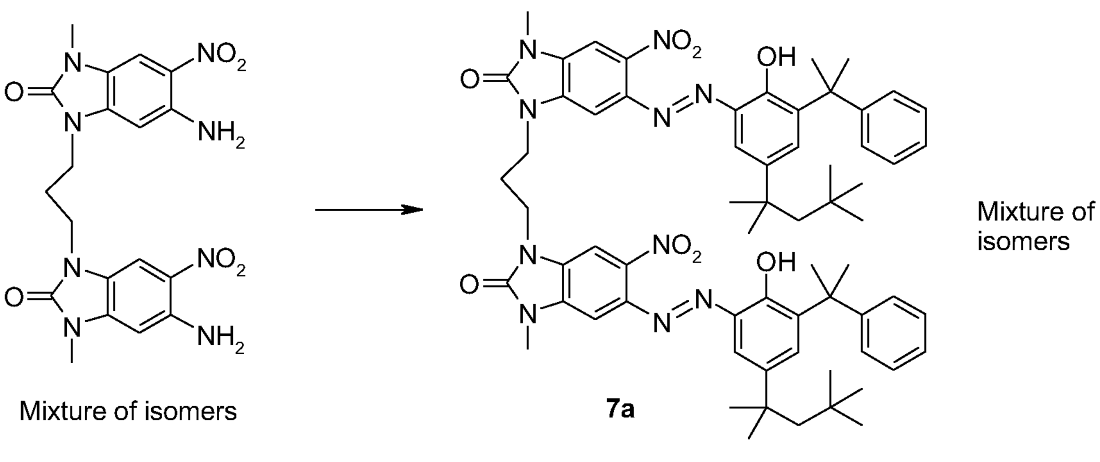
The above mentioned intermediate **7a** is prepared as follows:



A steel reactor is charged with 1,1'-(1,3-propanediyl)bis[3-methyl-5,6dinitrobenzimidazol-2-one]*) (10.3g; 20mmol) and 100ml of a 32% solution of ammonia in water. After stirring over night at 160°C (pressure rises up to 20bar) the reaction mixture is cooled to room temperature. The so formed diamino-dinitro-derivative is filtered off as an orange precipitate and washed with water.

Yield 7.9g (1.1mmol, 88%)

Melting point: 296°C decomposition



7a

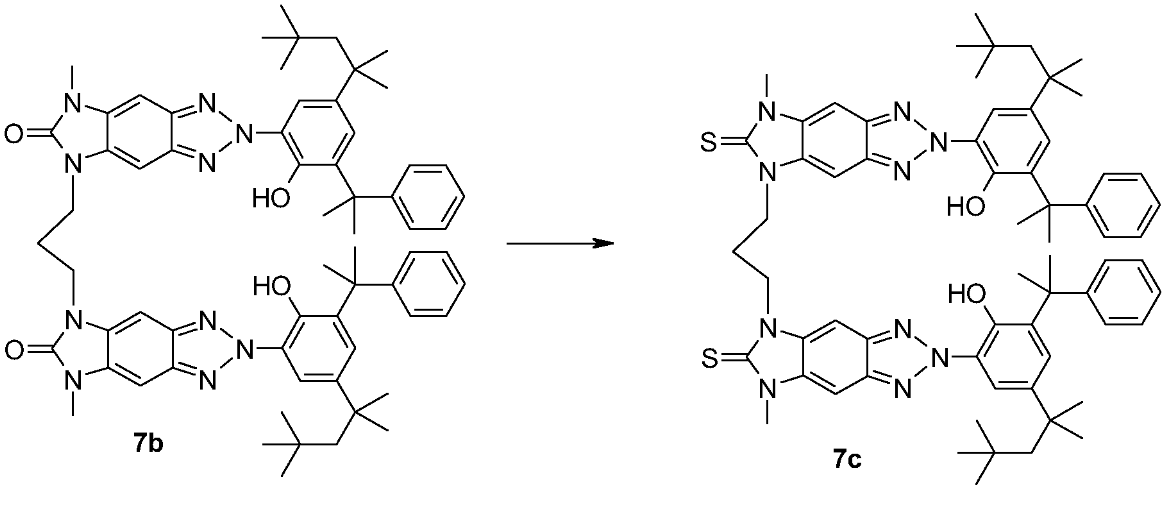
Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of the above described diaminodinitro derivate (6.9g, 15mmol) in acetic acid (70ml) and hydrochloric acid (32% in water; 10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (one hour), stirring is continued for 1.5 hours. The reaction mixture is then transferred into a dropping funnel and slowly added at -20°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 10.2g, 30mmol) in methanol / toluene (110ml; 6:5) containing sodium hydroxide microprills (1.2g, 30mmol). During the addition, the temperature of the reaction mixture is kept between -20°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held above 7 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hours; initial pH >11.0), the cooling bath is removed and the red suspension stirred overnight. After adding water (50ml) and toluene (100ml) The water phase is split off and washed with toluene (2x100ml). The combined organic phases are washed with water, dried (Na_2SO_4), filtered and the solvent evaporated.

Yield 19.3g (114%) crude

Melting point: 160-175°C (mixture of isomers)

*) 1,1'-(1,3-propanediyl)bis[3-methyl-5,6dinitrobenzimidazol-2-one] and corresponding o-nitro or o-chloro anilines can be prepared according to known procedures.

Example 10. Preparation of compound **7c**

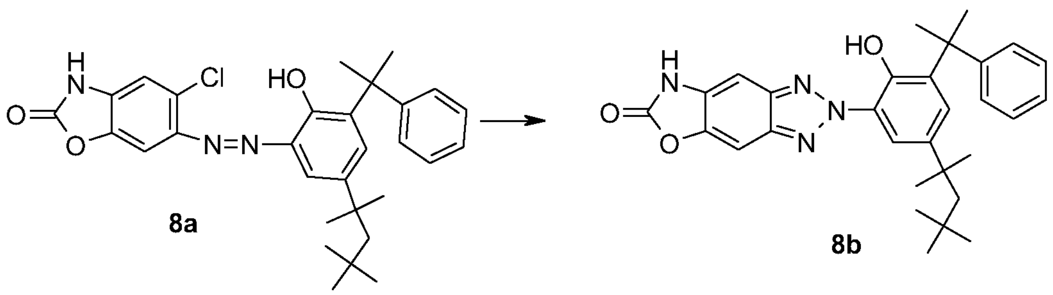


A stirred mixture of compound **7b** (2.4g, 2.25mmol), Lawesson Reagent (98%; 5.46g, 13.5mmol) and xylene (20ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled to room temperature, filtered and evaporated. Followed by chromatography on a silica gel column (hexane / ethyl acetate 7:3) and the obtained product is recrystallized from acetone.
Yield 1.2g (1.1mmol, 50%)

Melting point: 183°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 384 (98165)

Example 11. Preparation of compound **8b**

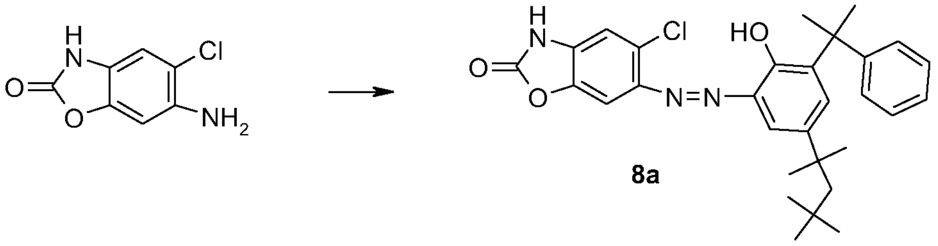


A stirred mixture of compound **8a** (17.8g; 34.2mmol), sodium azide (99%; 2.9g; 44.2mmol), copper(I) bromide (0.5g; 3.5mmol) and 1-methyl-2-pyrrolidinone (150ml) is heated to 120°C. The temperature is maintained and the progress of reaction is monitored by TLC (4 hours). The dark solution is added in a stirred mixture of ice and water. The residue is solved in ethyl actetate (150ml). The water phase is split off and washed with ethyl acetate (1x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 9:1)
Yield 10.7g (21.5mmol, 63%)

Melting point: 203°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 355 (22973)

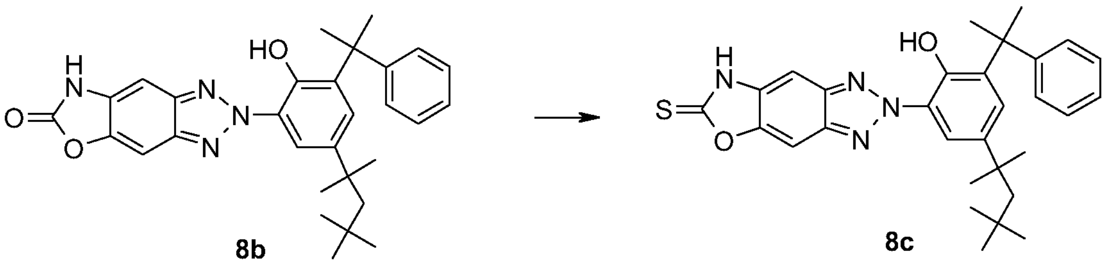
The above mentioned intermediate **8a** is prepared as follows:


8a

Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of 6-amino-5-chloro-benzoxazol-2-one (5g, 27mmol) in acetic acid (50ml) containing hydrochloric acid (32% in water; 15ml) and water (10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 8.8g, 27mmol) in methanol / xylene (120ml 85:15) containing sodium acetate (2.2g, 27mmol). During the addition, the temperature of the reaction mixture is kept between -20°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 8 and 9 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. After adding water (200ml) and ethyl acetate (300ml), the water phase is split off and washed with ethyl acetate (3x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is suspended with hot methanol. The cold suspension is filtered off, washed with methanol and dried. Yield 7.2g (13.8mmol, 51%).

Melting point: 200°C

Example 12. Preparation of compound **8c**

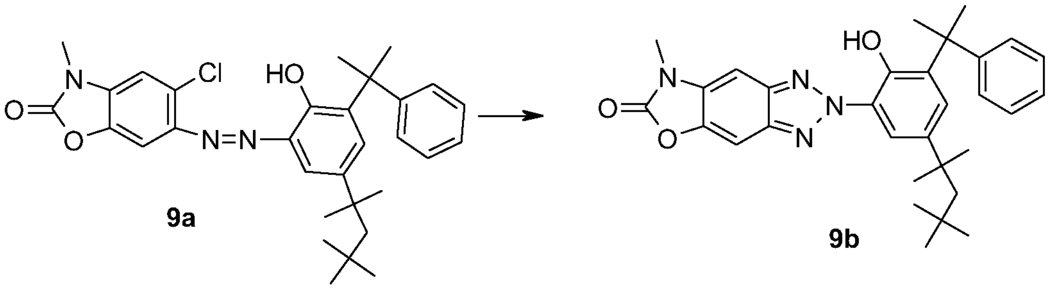


A stirred mixture of compound **8b** (1.0g, 2mmol), Lawesson Reagent (98%; 3.2g, 7.9mmol) and xylene (30ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled and filtered. The liquid layer is concentrated to dryness and the residue is chromatographed on a silica gel column (hexane / ethyl acetate 7:3)
Yield 0.8g (1.4mmol, 81%)

Melting point: 109°C.

UV-vis ($CHCl_3$ λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 375 (44920)

Example 13. Preparation of compound **9b**



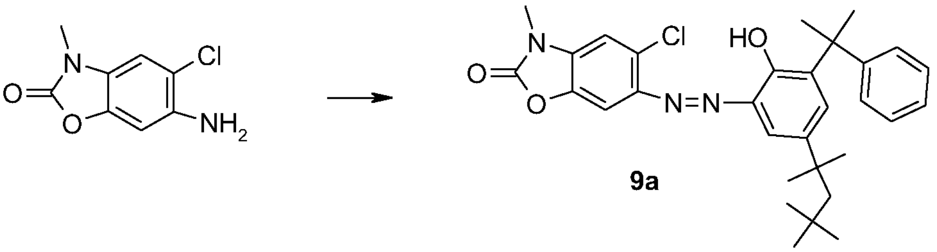
A stirred mixture of compound **9a** (4.0g; 7.5mmol), sodium azide (99%; 2.9g; 44.2mmol), copper(I) bromide (0.11g; 0.75mmol) and 1-methyl-2-pyrrolidinone (50ml) is heated to 120°C. The temperature is maintained and the progress of reaction is monitored by TLC (2 hours). The dark solution is added tp a stirred mixture of ice and water. The residue is dissolved in ethyl actetate (150ml). The water phase is split off and washed with ethyl acetate (1x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 7:3)

Yield 2.2g (4.3mmol, 57%)

Melting point: 176°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 357 (25423)

The above mentioned intermediate **9a** is prepared as follows:

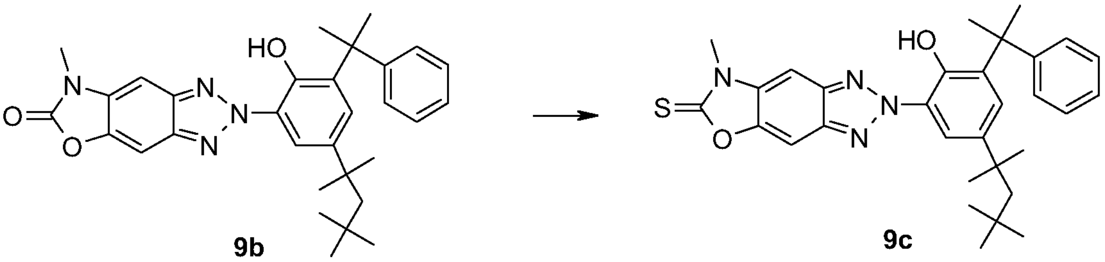


Sodium nitrite (4 molar in water; 8ml, 32mmol) is slowly added between 0°C and 5°C to a stirred solution of 6-amino-5-chloro-3-methyl-benzoxazol-2-one (5.8g, 29mmol) in acetic acid (60ml), hydrochloric acid (32% in water; 15ml) and water (10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 9.4g, 27.5mmol) in methanol / xylene (100ml 85:15) containing sodium acetate (2.4g, 29mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 6 and 8 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. After adding brine (200ml) and ethyl acetate (300ml), the water phase is split off and washed with ethyl acetate (3x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 6:2)

Yield 10.9g (20.4mmol, 70.4%)

Melting point: 231°C

Example 14. Preparation of compound **9c**



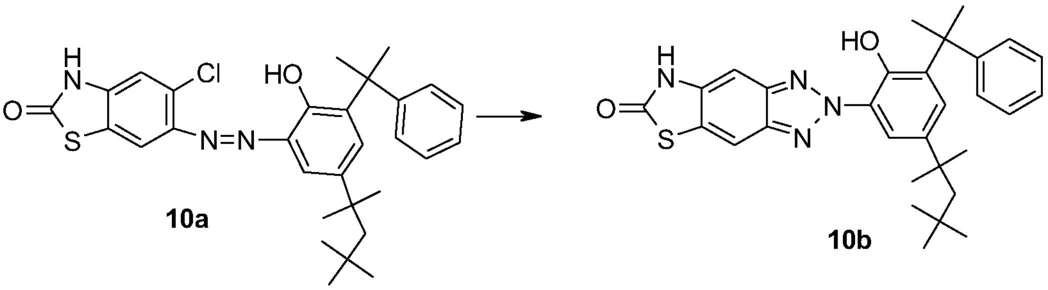
A stirred mixture of compound 9**b** (1.0g, 2mmol), Lawesson Reagent (98%; 3.2g, 7.9mmol) and xylene (30ml) is heated to 120°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled and filtered. The liquid layer is concentrated to dryness and the residue is crystallized with methanol.

Yield 0.8g (1.5mmol, 75%)

Melting point: 174°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / dm^3 mol^{-1} cm^{-1}): 377 (43647)

Example 15. Preparation of compound **10b**



A stirred mixture of compound **10a** (10.0g; 18.7mmol), sodium azide (99%; 1.6g; 24.2mmol), copper(I) bromide (0.27g; 1.9mmol) and 1-methyl-2-pyrrolidinone (50ml) is heated to 125°C.

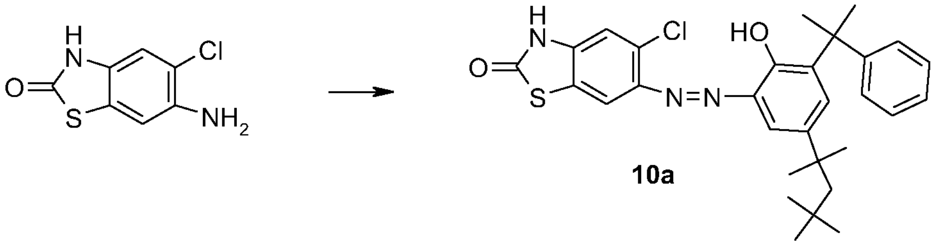
The temperature is maintained and the progress of reaction is monitored by TLC (8 hours). The dark solution is added in a stirred mixture of ice and water. The residue is solved in ethyl actetate (150ml). The water phase is split off and washed with ethyl acetate (1x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (cyclohexane / ethyl acetate 8:2)

Yield 3.3g (6.4mmol, 34%)

Melting point: 222°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 366 (27881)

The above mentioned intermediate **10a** is prepared as follows:

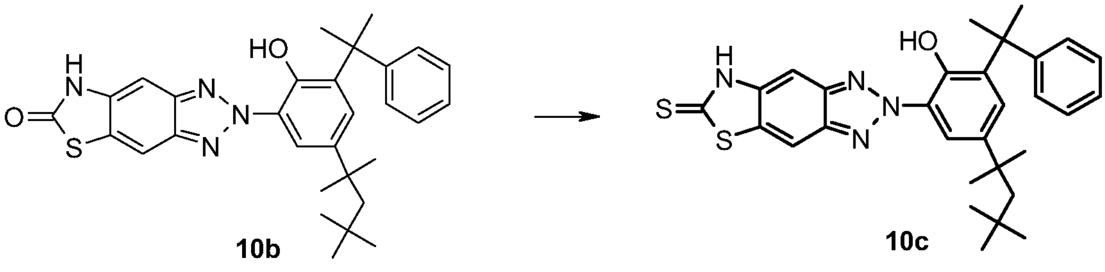


10a

Sodium nitrite (4 molar in water; 15ml, 60mmol) is slowly added between 0°C and 5°C to a stirred solution of 6-amino-5-chloro-benzothiazol-2-one (12.0g, 60mmol) in acetic acid (100ml) containing hydrochloric acid (32% in water; 50ml) and water (20ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (60 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (95%; 20.5g, 60mmol) in methanol / xylene (150ml 85:15) containing sodium acetate (4.9g, 60mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 7 and 9 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. After adding water (200ml) and ethyl acetate (500ml), the water phase is split off and washed with ethyl acetate (3x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is suspended with cold methanol (0°C). The cold suspension is filtered off, washed with cold methanol and dried. Yield 20.4g (38.1mmol, 64%).

Melting point: 253°C

Example 16. Preparation of compound **10c.**



10b

10c

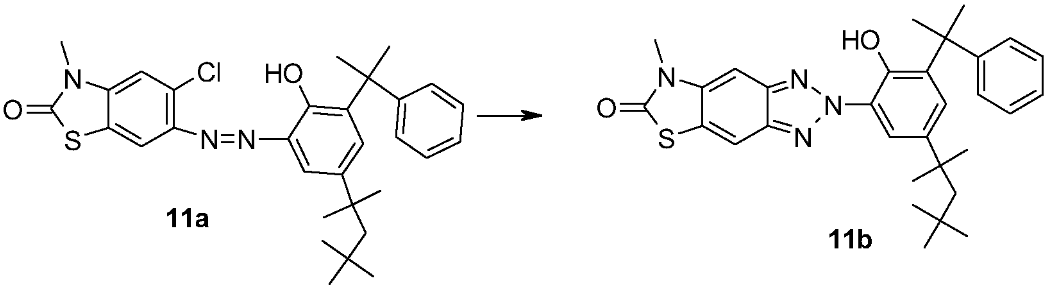
A stirred mixture of compound **10b** (2.4g, 4.7mmol), Lawesson Reagent (98%; 7.8g, 18.8mmol) and xylene (40ml) is heated to 120°C. The temperature is maintained and the progress of reaction is monitored by TLC (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled and filtered. The liquid layer is concentrated to dryness and the residue is chromatographed on a silica gel column (hexane / ethyl acetate 85:15)

Yield 1.3g (2.5mmol, 53%)

Melting point: 254°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 390 (46882)

Example 17. Preparation of compound **11b**



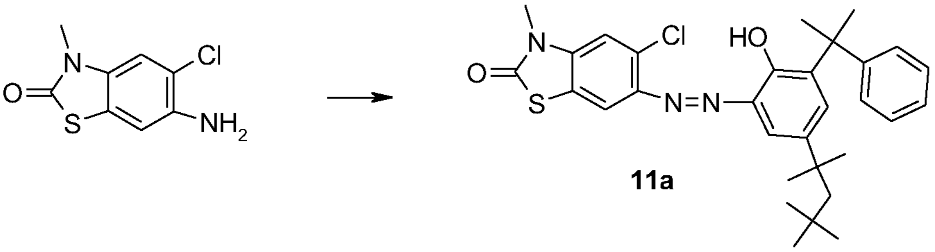
A stirred mixture of compound **11a** (20.0g; 36.4mmol), sodium azide (99%; 3g; 46.2mmol), copper(I) bromide (0.27g; 1.9mmol) and 1-methyl-2-pyrrolidinone (250ml) is heated to 125°C. The temperature is maintained and the progress of reaction is monitored by TLC (4.5 hours). The dark solution is added in a stirred mixture of ice and water (1000ml). The residue is filtered off and chromatographed on a silica gel column (cyclohexane / ethyl acetate 9.5:0.5)

Yield 16.1g (30.5mmol, 84%)

Melting point: 140°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 367 (27752)

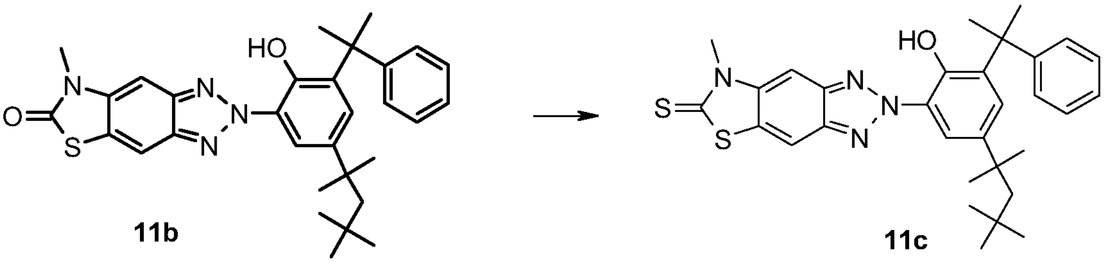
The above mentioned intermediate **11a** is prepared as follows:



Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of 6-amino-5-chloro-3-methyl-benzothiazol-2-one (6.7g, 31.2mmol) in acetic acid (60ml) containing hydrochloric acid (32% in water; 20ml) and water (10ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (45 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -10°C to a stirred solution of 2-cumyl-4-t-octylphenol (11.5g, 35mmol) in methanol / xylene (120ml; 85:15) containing sodium acetate (2.9g, 35mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 8 and 9 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. After adding brine (100ml) and ethyl acetate (300ml), the water phase is split off and washed with ethyl acetate (3x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is suspended with cold methanol (0°C). The cold suspension is filtered off, washed with cold methanol and dried. Yield 10.9g (19.8mmol, 57%).

Melting point: 255°C

Example 18. Preparation of compound **11c.**



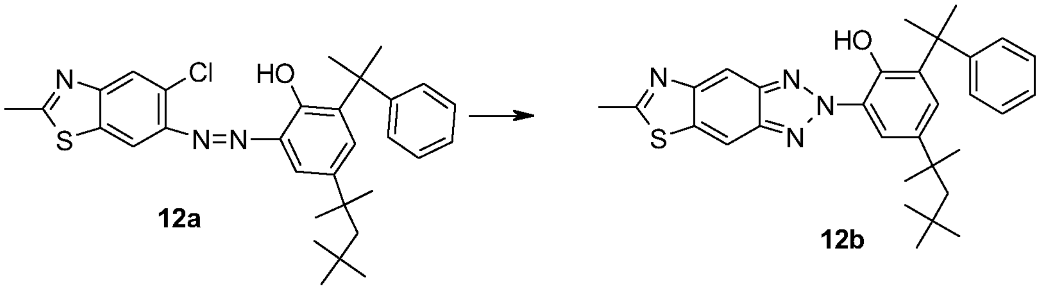
A stirred mixture of compound **11b** (1.31g, 2.5mmol), Lawesson Reagent (98%; 4.04g, 10mmol) and xylene (15ml) is heated to 120°C. The temperature is maintained and the progress of reaction is monitored by TLC (2 hours) (if necessary additional Lawesson Reagent is added). The reaction mixture is cooled and filtered. The liquid layer is concentrated to dryness and the residue is chromatographed on a silica gel column (hexane / ethyl acetate 95:5)

Yield 1.15g (2.1mmol, 84%)

Melting point: 120°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 392 (46042)

Example 19. Preparation of compound **12b**

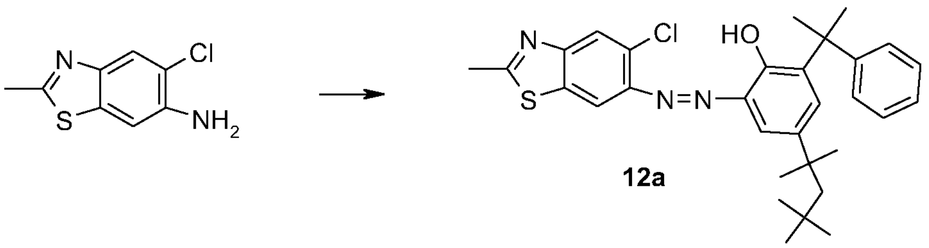


A stirred mixture of compound **12a** (5.35g; 10mmol), sodium azide (99%; 0.85g; 13mmol), copper(I) bromide (0.19g; 1.3mmol) and 1-methyl-2-pyrrolidinone (100ml) is heated to 60°C. The temperature is maintained and the progress of reaction is monitored by TLC (1.5 hours). The dark solution is added in water (100ml) and ethyl acetate (250ml). The water phase is split off and washed with ethyl acetate (2x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (hexane / ethyl acetate 4:1) and crystallized with hexane.
Yield 2.65g (5.2mmol, 52%)

Melting point: 188°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 364 (22533)

The above mentioned intermediate **12a** is prepared as follows:

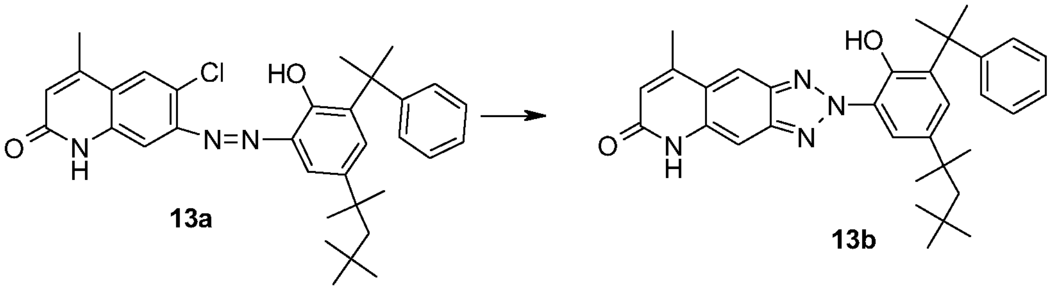


Sodium nitrite (4 molar in water; 10ml, 40mmol) is slowly added between 0°C and 5°C to a stirred solution of 5-chloro-2-methyl-benzothiazol-6-ylamine (7.2g, 31.2mmol) in acetic acid

(50ml) containing hydrochloric acid (32% in water; 12ml) and water (25ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (45 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -10°C to a stirred solution of 2-cumyl-4-t-octylphenol (11.5g, 35mmol) in methanol / xylene (120ml; 85:15) containing sodium acetate (4.7g, 56mmol) and sodium hydroxide microprills (1.4g; 35mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 7 and 9 by concomitant addition of sodium hydroxide (30% in water; 100ml). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. After adding brine (100ml) and dichloromethane (300ml), the water phase is split off and washed with dichloromethane (2x50ml). The combined organic phases are washed with brine, dried (Na_2SO_4), filtered and the solvent evaporated. The residue is recrystallized from methanol (500ml). Yield 12.4g (23.2mmol, 66%).

Melting point: 206°C

Example 20. Preparation of compound **13b**



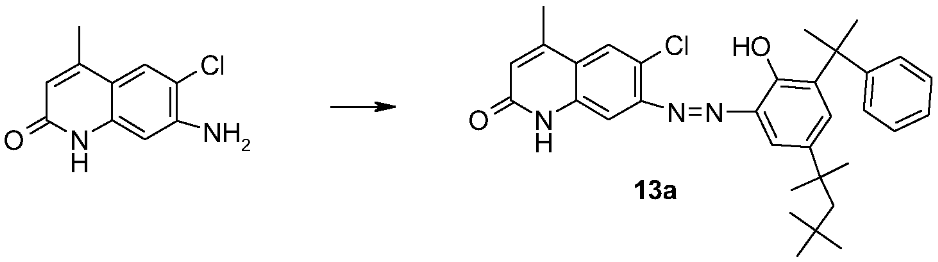
A stirred mixture of compound **13a** (5.45g; 10mmol), sodium azide (99%; 0.85g; 13mmol), copper(I) bromide (0.23g; 1.6mmol) and methyl sulfoxide (20ml) is heated to 70°C. The temperature is maintained and the progress of reaction is monitored by TLC (0.5 hours). The dark solution is added in water (200ml) and filtered off. The residue is suspended in hot isopropanol (25ml; 0.5 hours). After the suspension is cooled, the residue is filtered off and dryed.

Yield 4.6g (8.7mmol, 87%)

Melting point: 300°C.

UV-vis (dioxane), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 398 (40539), 419 (33575)

The above mentioned intermediate **13a** is prepared as follows:

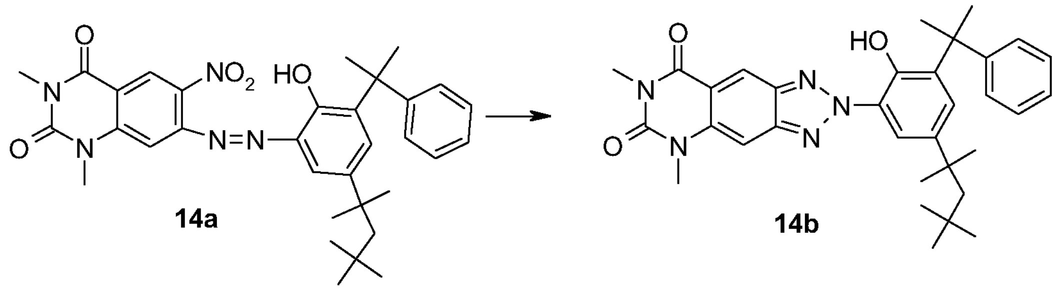


13a

Sodium nitrite (4 molar in water; 25ml, 100mmol) is slowly added between 0°C and 5°C to a stirred solution of 5-amino-6-chloro-4-methyl-2-chinolone (20.9g, 100mmol) in acetic acid (150ml) containing hydrochloric acid (37% in water; 35ml) and water (150ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued for 1 hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (32.5g, 100mmol) in methanol / xylene (200ml; 85:15) containing sodium acetate (13.2g, 160mmol) and sodium hydroxide microprills (2.0g; 50mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode and held between 7 and 12 by concomitant addition of sodium hydroxide (30% in water; 150ml). After the addition is complete (1 hour), the cooling bath is removed and the suspension stirred overnight. The residue is filtered off, washed with water and dried.
Yield 44.3g (84.5mmol, 85%).

Melting point: 299°C

Example 21. Preparation of compound **14b**



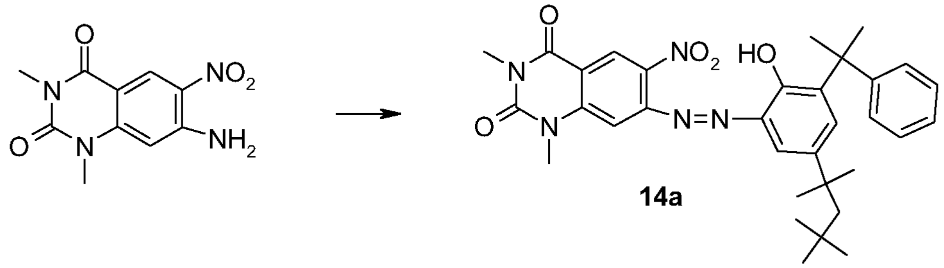
A stirred mixture of compound **14a** (16.6g; 28mmol), sodium azide (99%; 2.4g; 39mmol) and 1-methyl-2-pyrrolidinone (100ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (16 hours). The dark solution is poured onto ice (200g), extracted with toluene (2x100ml) and ethyl acetate (3x100ml) and the collected organic phases are evaporeted. The resulting 25g of red oil are chromatographed on a silica gel column (ethyl acetate / hexane 2:8) to yield 3.6 g of a solid which is purified again by semi preparative LC.

Yield 1.6g (2.9mmol, 10%)

Melting point: 209°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 391 (20670)

The above mentioned intermediate **14a** is prepared as follows:

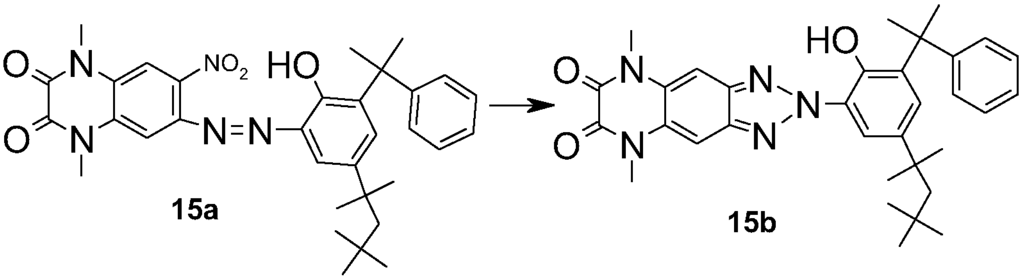


7-amino-1,3-dimethyl-6-nitro-quinazoline-2,4-dione (7.0g, 28mmol) are dissolved in concentrated sulfuric acid (40ml) and added dropwise to nitrosylsulfuric acid (40% in sulfuric acid; 9.8g, 31mmol). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued for another hour. The reaction mixture is then transferred into a dropping funnel and slowly added at -10°C to -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (9.1g, 28mmol) in methanol / m-xylene (50 / 50 by volume; 200ml) containing sodium hydroxide microprills (1.15g, 28mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -5°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode (initial pH >11) and later held between 5 and 8 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete the cooling bath is removed and the suspension stirred overnight. Water (100ml), brine (100ml) and ethyl acetate (250ml) are added, the water phase split off and washed with ethyl acetate (2x250ml). The combined organic phases are washed with brine (200ml), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (ethyl acetate / hexane 2:8).

Yield 24.0g (6.8mmol, 24%).

Melting point: 194°C decomposition

Example 22. Preparation of structure **15b**



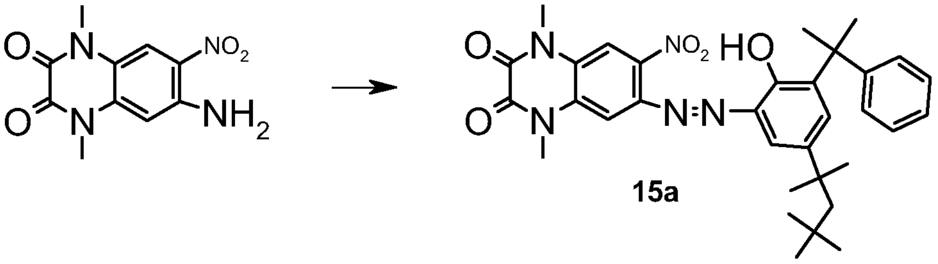
A stirred mixture of compound **15a** (6.0g; 10mmol), sodium azide (99%; 0.98g; 15mmol) and 1-methyl-2-pyrrolidinone (60ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (3 hours). After pouring the reaction mixture onto ice (100g), the crude product is filtered off and recrystallised from 1-methyl-2-pyrrolidinon.

Yield 1.6g (2.9mmol, 29%)

Melting point: 271°C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 382 (36555)

The above mentioned intermediate **15a** is prepared as follows:



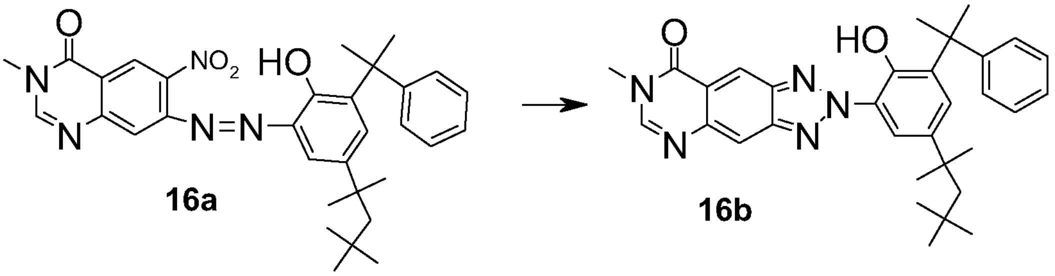
Nitrosylsulfuric acid (40% in sulfuric acid; 5.6g, 18mmol) is added dropwise to a solution of 6-amino-1,4-dimethyl-7-nitro-1,4-dihydro-quinoxaline-2,3-dione (4.0g, 16mmol) in concentrated sulfuric acid (20 ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued over night at room temperature. The reaction mixture is cooled again with an ice bath, diluted with 25g ice and then transferred into a dropping funnel and slowly added at -10°C to -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (5.5g, 16mmol) in methanol / m-xylene (50 / 50 by volume; 200ml) containing sodium hydroxide microprills (0.7g, 16mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -10°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode (initial pH > 11) and later held between 5 and 8 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete the cooling bath is removed and the suspension stirred overnight. Water (100ml), brine (100ml) and ethyl acetate (250ml) are added, the water phase split off and washed with ethyl acetate (2x250ml). The combined organic phases are washed with brine (200ml), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (ethyl acetate / hexane 1:1).

Yield 4.7g (8.0 mmol, 50%).

Melting point: 190°C decomposition

The mentioned ortho-nitroaniline can be prepared according to known procedures.

Example 23. Preparation of structure **16b**



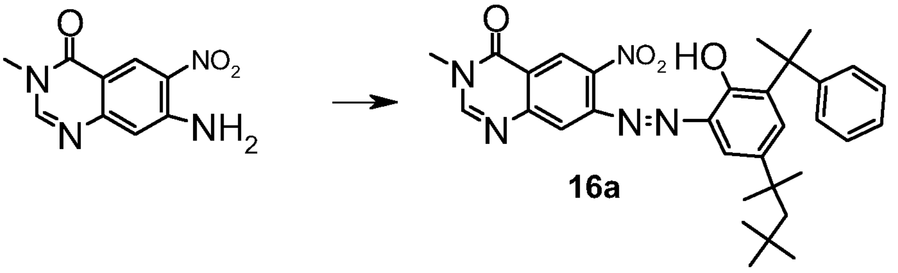
A stirred mixture of compound **16a** (0.8g; 1.4mmol), sodium azide (99%; 0.13g; 2.0mmol) and 1-methyl-2-pyrrolidinone (20ml) is heated to 130°C. The temperature is maintained and the progress of reaction is monitored by TLC (5 hours). The dark solution is poured onto ice (50g), extracted with ethyl acetate (3x20ml), the combined organic phases are washed with brine (2x30 ml), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (ethyl- acetate/hexane 7/3) to yield 0.43 g solid which is purified again by semi preparative LC.

Yield 0.06g (0.1mmol, 8%)

Melting point: 208 °C.

UV-vis ($CHCl_3$), λ_{max} / nm (ε / $dm^3 mol^{-1} cm^{-1}$): 388 (20880)

The above mentioned intermediate **16a** is prepared as follows:



Nitrosylsulfuric acid (40% in sulfuric acid; 3.5g, 11mmol) is added dropwise to a solution of 7-amino-3-methyl-6-nitro-3*H*-quinazolin-4-one (2.0g, 10mmol) in concentrated sulphuric acid

(30ml). During the addition the temperature of the reaction mixture is kept between 0°C and 5°C by means of an ice bath. After the addition is complete (30 minutes), stirring is continued for another two hours at room temperature. Then the reaction mixture is cooled again with an ice bath, diluted with 25 g ice, transferred into a dropping funnel and slowly added at -10°C to -15°C to a stirred solution of 2-cumyl-4-t-octylphenol (3.4g, 10mmol) in methanol / m-xylene (50 / 50 by volume; 100ml) containing sodium hydroxide micropills (0.6g, 15mmol) and sodium acetate (8g, 100mmol). During the addition, the temperature of the reaction mixture is kept between -15°C and -10°C by means of an isopropanol-dry ice bath; the pH is measured by means of an electrode (initial pH >11) and later held between 5 and 8 by concomitant addition of sodium hydroxide (30% in water). After the addition is complete the cooling bath is removed and the suspension stirred overnight. Water (100ml), brine (100ml) and ethyl acetate (200ml) are added, the water phase split off and washed with ethyl acetate (2x100ml). The combined organic phases are washed with brine (2x100ml), dried (Na_2SO_4), filtered and the solvent evaporated. The residue is chromatographed on a silica gel column (ethyl acetate / hexane 1:1).

Yield 0.32g (0.6mmol, 6%)

Melting point: 236°C decomposition

The mentioned ortho-nitroaniline can be prepared according to known procedures.

B) Application Examples

Materials used

Acryl/Melamine Clearcoat formulation:

a) Viacryl® SC 303[1] (65% solution in xylene/butanol, 26:9 wt./wt.)	27.51g
b) Viacryl® SC 370[2] (75% in Solvesso 100[3]	23.34g
c) Maprenal® MF 650[4] (55% in isobutanol)	27.29g
d) Butylacaetate / butanol (37:8 wt./wt.)	4.33g
e) Isobutanol	4.87g
f) Solvesso® 150[5]	2.72g
g) Crystal oil 30[6]	8.74g
h) Baysilone® MA[7] (1% in Solvesso® 150)	1.20g

Total	100.00g

Binder raw materials:

[1] Viacryl® SC 303: acrylic resin (Solutia, formerly Vianova Resins)

[2] Viacryl® SC 370: acrylic resin (Solutia, formerly Vianova Resins)

[3] Solvesso® 100: aromatic hydrocarbon, bp. 163-180°C (Exxon Corp.)

[4] Maprenal® MF 650: melamine resin (Solutia, formerly Vianova Resins)

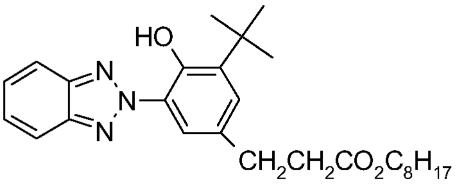
[5] Solvesso® 150: aromatic hydrocarbon, bp. 180-203°C (Exxon Corp.)

[6] Crystal oil 30: aliphatic hydrocarbon, bp. 145- 200°C (Shell Corp.)

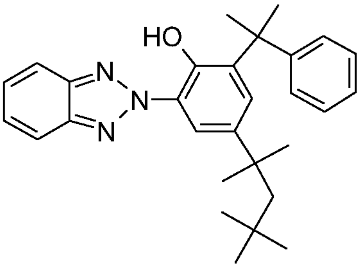
[7] Baysilone® MA: leveling agent (Bayer AG)

UV-absorbers and other stabilizers

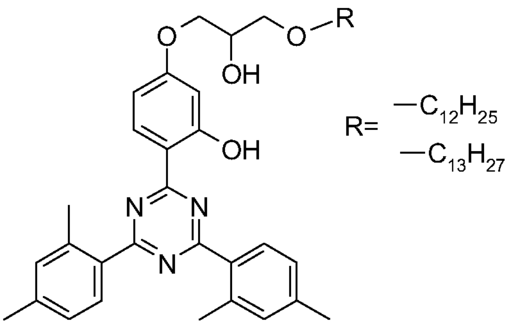
Tinuvin 384® (commercial UV-Absorber from Ciba Specialty Chemicals)



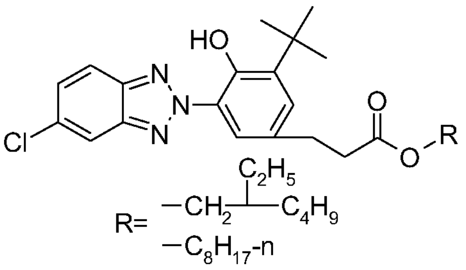
Tinuvin 928® (commercial UV-absorber from Ciba Specialty Chemicals)



Tinuvin 400® (commercial UV-absorber from Ciba Specialty Chemicals)

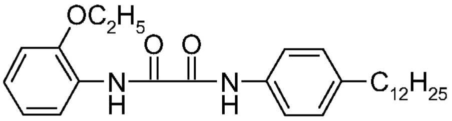


Tinuvin 109®



(commercial UV-absorber from Ciba Specialty Chemicals)

Oxanilide-1



Compounds 1b and 3d (I)-(V) of example A1 and A3 are UV-absorbers according to the invention.

Tinuvin 152® (hindered amine stabilizer from Ciba Specialty Chemicals)

Example B1: Photo permanence of UV-absorbers

The photo permanence of the UV-absorbers is evaluated as follows:
the UV-absorbers of the present invention are incorporated into a thermosetting acryl / melamine clear coat (based on Viacryl® SC 303 / Viacryl® SC 370 / Maprenal® MF 650) in a concentration of 3% based on the solids content of the formulation (solids content: 50.4%). The clear coat is sprayed onto glass plates resulting in a dry film thickness of the clear coat of 20µm after cure (130°C / 30').

Prior to exposure of the specimens, the UV-absorption spectra are recorded using a UV / VIS spectrometer (Perkin Elmer, Lamda 40). Reference: unstabilized acryl / melamine clear coat. Subsequently the specimens are exposed in a Xenon – WOM wetherometer (Atlas Corp.) according to SAE J 1960. The percentage of UV-absorber retained (determined at λ max.) upon exposure is monitored by recording the UV-absorption spectra after regular exposure intervals. The test results are summarized in Table 1:

Table 1: photo permanence of claimed UV-absorbers during Xe-WOM exposure in comparison to commercial references

Sample	% UV-absorber retained after … hours Xe-WOM exposure				
	1000	1500	2000	3000	4000h
compound **14b**	94.6	92.4	83.2	73.3	63.1
hydroxy-phenyl – benzotriazole [1)]	68.3	50.8	40.2	18.7	n.a.
hydroxy-phenyl-benzotriazole [2)]	83.0	n.a.	68.2	43.4	n.a
hydroxy-phenyl-triazine [3)]	89.8	82.1	76.0	59.4	n.a.
Oxanilide-1	8				

1) Tinuvin 384
2) Tinuvin 928
3) Tinuvin 400

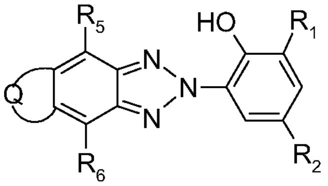
Example B 2:

In a second example two subsequent clear coats are applied on top of each other. The first clear coat (Clear coat I) is stabilized and applied as described in greater detail in example 1. A second thermosetting acryl / melamine clear coat (based on Viacryl® SC 303 / Viacryl® SC 370 / Maprenal® MF 650) is subsequently sprayed onto the first clear coat resulting in a dry film thickness of the second clear coat (clear coat II) of 40µm after cure (130°C / 30'). The second clear coat is stabilized using a UV - absorber combination of 3% Tinuvin 109 / 1.5% Tinuvin 400 and 1% Tinuvin 152 as co-stabilizer (HALS). Reference: unstabilized first clear coat. As described in example 1, the UV-transmission spectra are recorded prior to exposure of the specimens using a UV / VIS spectrometer (Perkin Elmer, Lamda 40). Subsequently the specimens are exposed in a Xenon – WOM wetherometer (Atlas Corp.) according to SAE J 1960. The transmission values (determined at 396 nm) as a function of the exposure period are monitored by recording the transmission spectra after regular exposure intervals. The test results are summarized in Table 2:

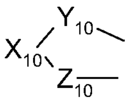
Table 2: transmission values (determined at 396nm) as a function of exposure intervals during Xe-WOM exposure

Sample	transmission values (%) after …. hours				
	initial	1000	2000	3000	4000
Clear coat I: **unstabilized** Clear coat II: 3% Tinuvin 109 / 1.5% Tinuvin 400	21.3	24.3	25.4	26.4	28.1
Clear coat I: **3%** compound **14b** Clear coat II: 3% Tinuvin 109 / 1.5% Tinuvin 400	0.35	0.38	0.46	0.62	0.74

Claims

1. A compound of formula (I)  (I) wherein

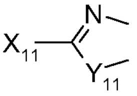
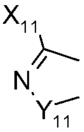
Q is a heterocyclic radical forming a 5 or 6-membered ring together with the annealed phenyl-ring, which radical is selected from the group consisting of

 wherein

X_{10} is O=C, S=C, $R_{101}N$, $S(O)_n$ where n is 0, 1 or 2;

Y_{10} is NR_{101}, O, $S(O)_n$ where n is 0, 1 or 2;

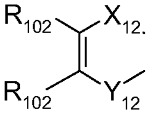
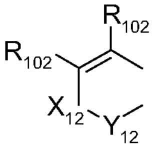
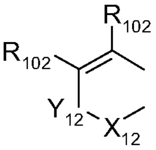
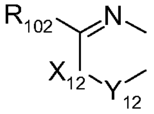
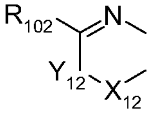
Z_{10} is O=C, S=C, $R_{101}N$, O, $S(O)_n$ where n is 0, 1 or 2;

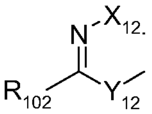
 , 

wherein

X_{11} is R_{101}, $N(R_{101})_2$, OR_{101}, $S(O)_nR_{101}$ where n is 0, 1 or 2;

Y_{11} is C=O, NR_{101}, O, or $S(O)_n$ where n is 0, 1 or 2;

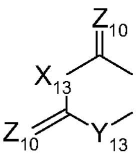
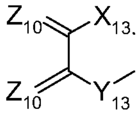
 ,  ,  ,  ,  ,



wherein

X_{12} is C=O or C=S;

Y_{12} is NR_{101}, O or S;

 , and 

wherein

X_{13} and **Y_{13}** are independently NR_{101}, O or S;

the **Z_{10}** are independently from each other O or S;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more -halogen, -OH, -OR_{22}, -NH_2, -NHR_{22}, -$N(R_{22})_2$, -$NHCOR_{23}$, -$NR_{22}COR_{23}$, -$OCOR_{24}$, -COR_{25}, -SO_2R_{26}, -$SO_3^-M^+$, -$PO(R_{27})_n(R_{28})_{2-n}$, -$Si(R_{29})_n(R_{30})_{3-n}$, -$Si(R_{22})_3$, -$N^+(R_{22})_3$ A^-, -$S^+(R_{22})_2$ A^- or combinations thereof; said straight or branched chain unsubstituted or substituted C_1-C_{24} alkyl, straight or branched chain unsubstituted or substituted C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

said phenyl, naphthyl or C_7-C_{15}phenylalkyl can also be substituted by one ore more -halogen, -CN, -CF_3, -NO_2, -NHR_{22}, -$N(R_{22})_2$, -SO_2R_{26}, -$PO(R_{27})_n(R_{28})_{2-n}$, -OH, -$OR_{22}$, -$COR_{25}$, -$R_{25}$; wherein

n is 0, 1 or 2;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl, C_7-C_{15} phenylalkyl, or two R_{22} when attached to an N or Si atom can form together with the atom to which they are bonded a pyrrolidine, piperidine or morpholine ring;

R_{23} is hydrogen, OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{25} is hydrogen, OH, OR_{22}, NHR_{22} or $N(R_{22})_2$, or has the same meaning as R_{22},

R_{26} is OH, OR_{22}, NHR_{22} or $N(R_{22})_2$,

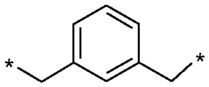
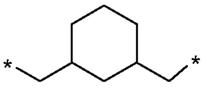
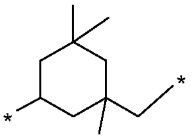
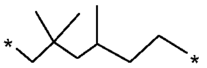
R_{27} is NH_2, NHR_{22} or $N(R_{22})_2$,

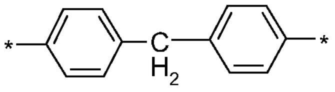
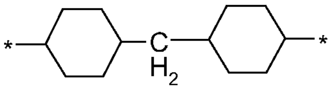
R_{28} is OH or OR_{22},

R_{29} is Cl or OR_{22},

R_{30} is straight or branched chain C_1-C_{18} alkyl; or

R_{101} can be a bridging group of straight or branched C_1-C_{18}alkylene, C_5-C_{10}cycloalkylene, para-phenylene or a group

 ,  ,  ,  ,

 ,  wherein * denotes a bond

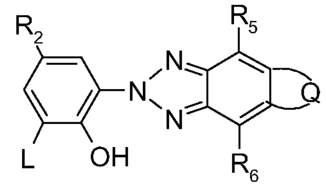
where the bridge connects two compounds of formula I, said C_1-C_{12}alkylene, C_5-C_{10}cycloalkylene can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

or when Y is a direct bond, Z can additionally also be a direct bond;

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or

if two substituents R_{101}/R_{101}, R_{101}/R_{102} or R_{102}/R_{102} are in vicinal position, they can form together with the atoms to which they are bonded an aliphatic, 5 to 8-membered ring;

R_1 is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms; or

R_1 is a group  wherein

L is alkylene of 1 to 12 carbon atoms, alkylidene of 2 to 12 carbon atoms, benzylidene, p-xylylene or cycloalkylene of 5 to 7 carbon atoms;

R_2 is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

said alkyl substituted by one or more -OH, -OCO-R_{11}, -OR_{14}, -NCO or -NH_2 groups or mixtures thereof, or said alkyl or said alkenyl interrupted by one or more -O-, -NH- or –NR_{14}-

groups or mixtures thereof and which can be unsubstituted or substituted by one or more -OH, -OR$_{14}$ or -NH$_2$ groups or mixtures thereof; where

R$_{11}$ is hydrogen, straight or branched chain C$_1$-C$_{18}$alkyl, C$_5$-C$_{12}$cycloalkyl, straight or branched chain C$_3$-C$_8$alkenyl, phenyl, naphthyl or C$_7$-C$_{15}$phenylalkyl; and

R$_{14}$ is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms; or

R$_2$ is –OR$_{14}$, a group –C(O)-O-R$_{14}$, -C(O)-NHR$_{14}$ or –C(O)-NR$_{14}$R'$_{14}$ wherein R'$_{14}$ has the same meaning as R$_{14}$; or

R$_2$ is –SR$_{13}$, -NHR$_{13}$ or -N(R$_{13}$)$_2$; or

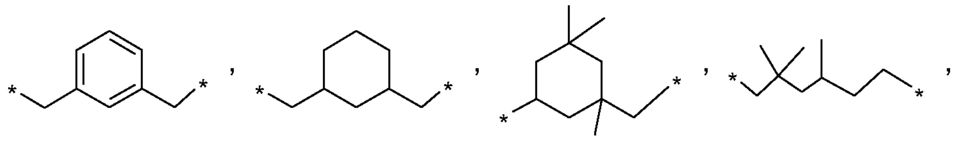
R$_2$ is -(CH$_2$)$_m$-CO-X$_1$-(Z)$_p$-Y-R$_{15}$ wherein

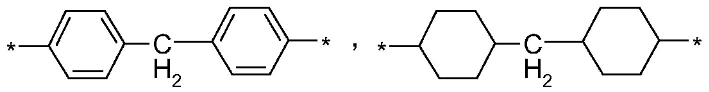
X$_1$ is -O- or -N(R$_{16}$)-,

Y is -O- or -N(R$_{17}$)- or a direct bond,

Z is C$_2$-C$_{12}$-alkylene, C$_4$-C$_{12}$alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or is C$_3$-C$_{12}$alkylene, butenylene, butynylene, cyclohexylene or phenylene, each of which may be additionally substituted by a hydroxyl group;

or a group





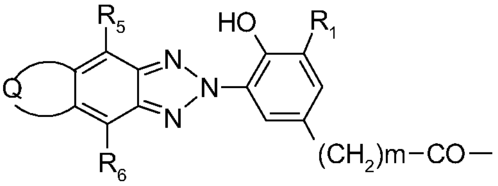
wherein * denotes a bond

or when Y is a direct bond, Z can additionally also be a direct bond;

m is zero, 1 or 2,

p is 1, or p is also zero when X and Y are -N(R$_{16}$)- and -N(R$_{17}$)-, respectively,

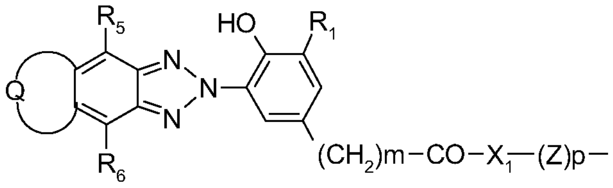
R$_{15}$ is hydrogen, C$_1$-C$_{12}$alkyl, a group



or a group -CO-C(R$_{18}$)=C(H)R$_{19}$ or, when Y is -N(R$_{17}$)-, forms together with R$_{17}$ a group -CO-CH=CH-CO- wherein

R$_{18}$ is hydrogen or methyl, and **R$_{19}$** is hydrogen, methyl or -CO-X$_1$-R$_{20}$, wherein

R$_{20}$ is hydrogen, C$_1$-C$_{12}$alkyl or a group of formulae



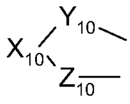
;

R_5 and R_6 are independently hydrogen or C_1-C_{18}alkylene;

R_{13} is alkyl of 1 to 20 carbon atoms, hydroxyalkyl of 2 to 20 carbon atoms, alkenyl of 3 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl or naphthyl, which both may be substituted by one or two alkyl of 1 to 4 carbon atoms;

R_{16} and **R_{17}** independently of one another are hydrogen, C_1-C_{12}-alkyl, C_3-C_{12}-alkyl interrupted by 1 to 3 oxygen atoms, or is cyclohexyl or C_7-C_{15}phenylalkyl, and R_{16} together with R_{17} in the case where Z is ethylene, also forms ethylene.

2. A compound of formula (I) according to claim 1 wherein Q is



Y_{10} is NR_{101}

X_{10} is O=C, S=C, $R_{101}N$;

Z_{10} is O=C, S=C, $R_{101}N$, O, $S(O)_n$ where n is 0;

or **Y_{10}** is O

X_{10} is O=C, S=C;

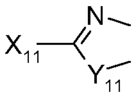
Z_{10} is $R_{101}N$;

or **Y_{10}** is S

X_{10} is O=C, S=C;

Z_{10} is $R_{101}N$;

or Q is



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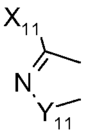
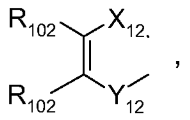


X_{11} is R_{101}, $N(R_{101})_2$, OR_{101}, $S(O)_nR_{101}$ where n is 0;

Y_{11} is C=O, NR_{101}, O, or $S(O)_n$ where n is 0;

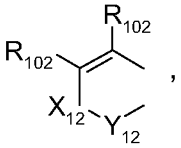
or Q is



X_{12} is C=O;

Y_{12} is NR_{101}, O or S;

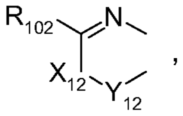
or Q is



X_{12} is C=O;

Y_{12} is NR_{101}, O or S;

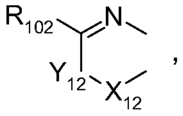
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

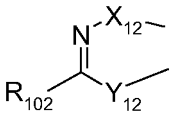
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

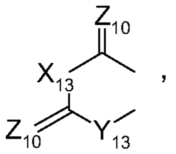
or Q is



X_{12} is C=O;

Y_{12} is NR_{101} or O;

or Q is

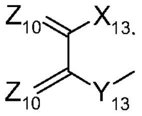


X_{13} is NR_{101}, O or S;

Y_{13} is NR_{101};

Z_{10} is O;

or Q is



X_{13} is NR_{101};

Y_{13} is NR_{101};

Z_{10} is O;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; or said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more -halogen, -OH, -OR_{22}, -NH_2, -NHR_{22}, -$N(R_{22})_2$, -$NHCOR_{23}$, -$NR_{22}COR_{23}$, -$OCOR_{24}$, -COR_{25}, -SO_2R_{26}, -$SO_3^-M^+$, -$PO(R_{27})_n(R_{28})_{2-n}$, -$Si(R_{29})_n(R_{30})_{3-n}$, -$Si(R_{22})_3$, -$N^+(R_{22})_3$ A^-, -$S^+(R_{22})_2$ A^- or combinations thereof; said straight or branched chain unsubstituted or substituted C_1-C_{24} alkyl, straight or branched chain unsubstituted or substituted C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

said phenyl, naphthyl or C_7-C_{15}phenylalkyl can also be substituted by one ore more -halogen, -CN, -CF_3, -NO_2, -NHR_{22}, -$N(R_{22})_2$, -SO_2R_{26}, -$PO(R_{27})_n(R_{28})_{2-n}$, -OH, -$OR_{22}$, -$COR_{25}$, -$R_{25}$; wherein

n is 0, 1 or 2;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl, C_7-C_{15} phenylalkyl, or two R_{22} when attached to an N or Si atom can form together with the atom to which they are bonded a pyrrolidine, piperidine or morpholine ring;

R_{23} is hydrogen, OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22},

R_{25} is hydrogen, OH, OR_{22}, NHR_{22} or $N(R_{22})_2$, or has the same meaning as R_{22},

R_{26} is OH, OR_{22}, NHR_{22} or $N(R_{22})_2$,

R_{27} is NH_2, NHR_{22} or $N(R_{22})_2$,

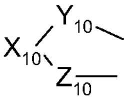
R_{28} is OH or OR_{22},

R_{29} is Cl or OR_{22},

R_{30} is straight or branched chain C_1-C_{18} alkyl; and

R_{102} is hydrogen, -CN, $-COR_{24}$ straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl.

3. A compound of formula (I) according to claim 2 wherein Q is



Y_{10} is NR_{101}

X_{10} is O=C, S=C

Z_{10} is O=C, $R_{101}N$, O, $S(O)_n$ where n is 0;

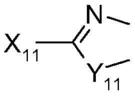
or

Y_{10} is NR_{101}

X_{10} is NR_{101}

Z_{10} is O=C;

or Q is



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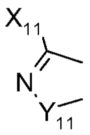
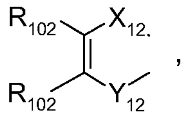


X_{11} is R_{101};

Y_{11} is C=O, NR_{101}, O, or $S(O)_n$ where n is 0;

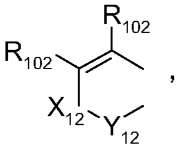
or Q is



X$_{12}$ is C=O;

Y$_{12}$ is NR$_{101}$ or O;

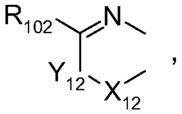
or Q is



X$_{12}$ is C=O;

Y$_{12}$ is NR$_{101}$ or O;

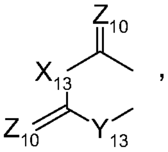
or Q is



X$_{12}$ is C=O;

Y$_{12}$ is NR$_{101}$ or O;

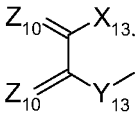
or Q is



X$_{13}$ is NR$_{101}$;

Y$_{13}$ is NR$_{101}$;

Z$_{10}$ is O;

or Q is 

X$_{13}$ is NR$_{101}$;

Y$_{13}$ is NR$_{101}$;

Z$_{10}$ is O;

R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; said straight or branched chain said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more –OH; or can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}-groups or combinations thereof;

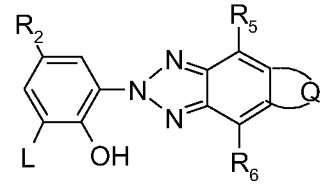
R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl or C_7-C_{15} phenylalkyl;

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; and

R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22}.

4. A compound of formula (I) according to claim 1 wherein

R_1 is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms; or

R_1 is a group 

wherein L is alkylene of 1 to 12 carbon atoms, alkylidene of 2 to 12 carbon atoms, benzylidene, p-xylylene or cycloalkylene of 5 to 7 carbon atoms;

R_2 is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

R_2 is -$(CH_2)_m$-CO-X_1-$(Z)_p$-Y-R_{15} wherein

X_1 is -O-,

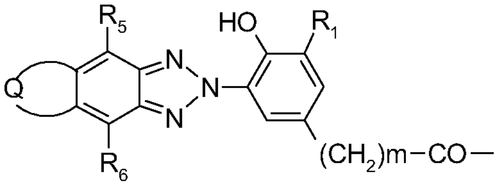
Y is -O- or a direct bond,

Z is C_2-C_{12}-alkylene, C_4-C_{12}alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or when Y is a direct bond, Z can additionally also be a direct bond;

m is 2,

p is 1,

R_{15} is hydrogen, C_1-C_{12}alkyl or a group



R_5 and R_6 are independently hydrogen or C_1-C_4alkyl.

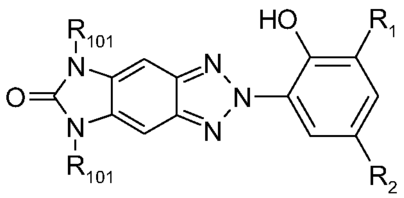
5. A compound of formula (I) according to claim 1 wherein

R_1 is hydrogen, straight or branched chain alkyl of 1 to 12 carbon atoms or phenylalkyl of 7 to 15 carbon atoms;

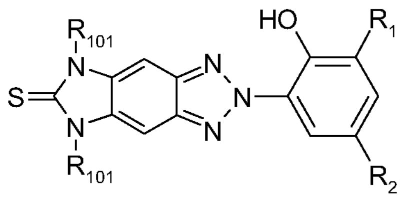
R_2 is straight or branched chain alkyl of 1 to 12 carbon atoms or phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms;

R_5 and **R_6** are hydrogen or C_1-C_4alkyl.

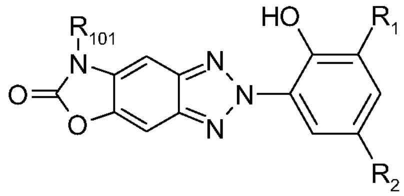
6. A compound of formula (I) according to claim 1 wherein the compound of formula (I) is a compound according to formulae (a) to (i)



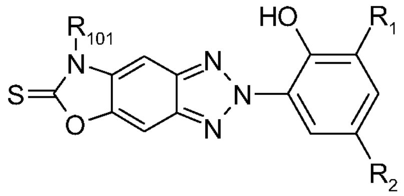
(a) ,



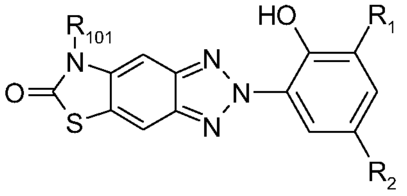
(b),

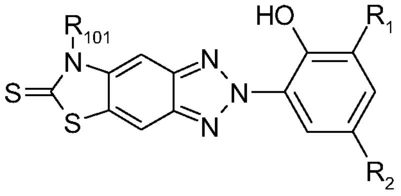


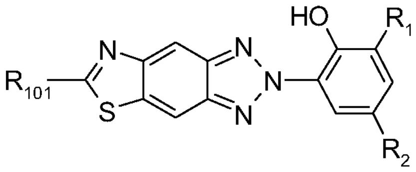
(c) ,

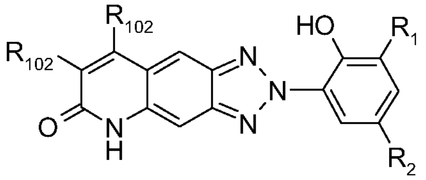


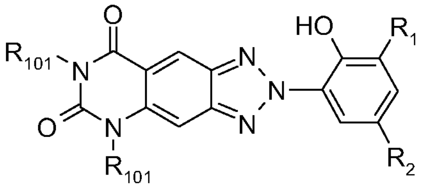
(d),

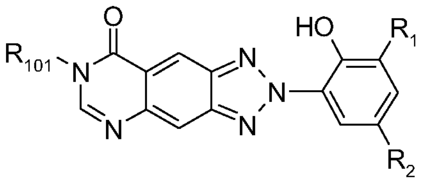

(e),

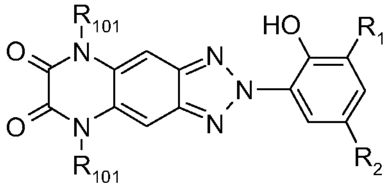

(f)


(g) ,


(h) and


(i) ,


or


wherein

R_1 is hydrogen, straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 4 alkyl of 1 to 4 carbon atoms;

R_2 is straight or branched chain alkyl of 1 to 24 carbon atoms, straight or branched chain alkenyl of 2 to 18 carbon atoms, cycloalkyl of 5 to 12 carbon atoms, phenylalkyl of 7 to 15 carbon atoms, phenyl, or said phenyl or said phenylalkyl substituted on the phenyl ring by 1 to 3 alkyl of 1 to 4 carbon atoms; or

R_2 is $-(CH_2)_m-CO-X_1-(Z)_p-Y-R_{15}$ wherein

X_1 is -O-,

Y is -O- or a direct bond,

Z is C_2-C_{12}-alkylene, C_4-C_{12}alkylene interrupted by one to three nitrogen atoms, oxygen atoms or a mixture thereof, or when Y is a direct bond, Z can additionally also be a direct bond;

m is 2,

p is 1,

R_{15} is hydrogen, C_1-C_{12}alkyl;

R_5 and R_6 are hydrogen;

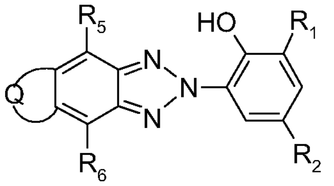
R_{101} is hydrogen, straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; said straight or branched chain C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl, C_2-C_6 alkinyl can be substituted by one or more –OH; or said straight or branched chain unsubstituted or substituted C_1-C_{24} alkyl, straight or branched chain C_2-C_{24} alkenyl, C_5-C_{12} cycloalkyl or C_2-C_6 alkinyl can also be interrupted by one or more -O-, -S-, -NH- or -NR_{22}- groups or combinations thereof;

R_{22} is straight or branched chain C_1-C_{18} alkyl, straight or branched chain C_2-C_{18} alkenyl, C_5-C_{10} cycloalkyl, phenyl or naphthyl or C_7-C_{15} phenylalkyl;

R_{102} is hydrogen, -CN, -COR_{24} straight or branched chain C_1-C_{24}alkyl, straight or branched chain C_2-C_{18}alkenyl, C_2-C_6alkyinyl, C_5-C_{12}cycloalkyl, phenyl, naphthyl or C_7-C_{15}phenylalkyl; and

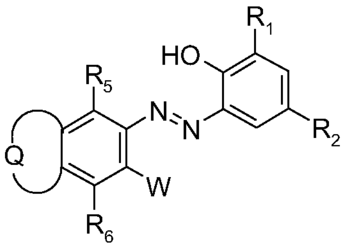
R_{24} is OR_{22}, NHR_{22}, $N(R_{22})_2$ or has the same meaning as R_{22}.

7. A process for the preparation of a compound of formula (I) according to claim 1



(I)

wherein the substituents R_1 to R_6 and Q are as defined in claim 1, which process comprises reacting a compound of formulae (III)

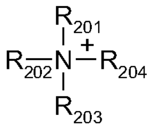


(III), wherein W is halogen or nitro

with an azide compound of formula (X)

$$M^{n+} (N_3^-)_r \qquad (X)$$

wherein

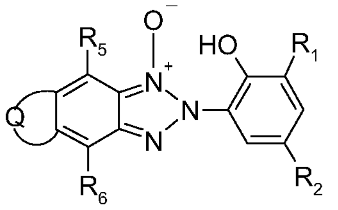
M is an n-valent metal cation,  $R_{202}-\overset{R_{201}}{\underset{R_{203}}{N^{+}}}-R_{204}$ or $P^{+}(R_{205})_4$,

$\mathbf{R_{201}}$, $\mathbf{R_{202}}$, $\mathbf{R_{203}}$ and $\mathbf{R_{204}}$ are each independently of the others hydrogen or C_1-C_{18}alkyl,
$\mathbf{R_{205}}$ is C_1-C_{18}alkyl, and
r is 1, 2 or 3.

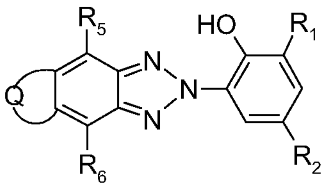
8. A process for the preparation of a compound of formula (I) according to claim 7 wherein the reaction is carried out in a solvent.

9. A process for the preparation of a compound of formula (I) according to claim 7 wherein the molar ratio of the amount of compound of formula III to the amount of azide compound of formula X is from 1 : 1 to 1 : 3.

10. A process according to claim 7, wherein the reaction is carried out in the presence of a catalyst.

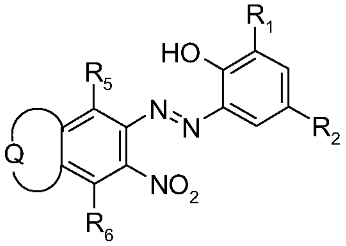
11. A compound of formula (IV)  (IV) wherein the substituents R_1 to R_6 and Q are as defined in claim 1.

12. A process for the preparation of a compound of formula (I) according to claim 1



(I)

wherein the substituents R_1 to R_6 and Q are as defined in claim 1, which process comprises reacting a compound of formulae (III)

 (III) under reductive conditions to a compound of formula (I).

13. A composition stabilized against light-induced degradation which comprises,
    (a) an organic material subject to light-induced degradation, and
    (b) a compound of formula I according to claim 1.

14. A composition according to claim 13 which contains additionally a sterically hindered amine stabilizer and/or a UV absorber selected from the group consisting of the s-triazines, the oxanilides, the hydroxybenzophenones, benzoates, the α-cyanoacrylates and the benzotriazoles different from those of formulae (I) according to claim 1.

15. A composition according to claim 13 wherein the organic material is a recording material.

16. A composition according to claim 13 wherein the organic material is a natural, semi-synthetic or synthetic polymer.

17. A composition according to claim 16 wherein the polymer is a thermoplastic polymer.

18. A composition according to claim 13 wherein the organic material is a coating.

19. A composition according to claim 18 wherein the coating is an automotive coating.

20. A composition according to claim 18 wherein the coating is applied over a substrate, which is sensitive to electromagnetic radiation of wavelengths greater than 380 nm.

21. A composition according to claim 19 wherein the automotive coating comprises
a) a primer coat which is electrodeposited onto a metal substrate;
b) at least one pigmented base coat which is in direct contact with the primer coat, containing a compound of formula (I) according to claim 1;
c) a clear coat which is in direct contact with the pigmented base coat, containing a UV-absorber selected from the group consisting of the s-triazines, the oxanilides, the

hydroxybenzophenones, benzoates, the α-cyanoacrylates and the benzotriazoles different from those of formulae (I) according to claim 1 and

d) optionally the primer coat comprises a compound of formula (I) according to claim 1.

22. A composition according to claim 21 which contains in the primer coat a compound of formula (I) according to claim 1.

23. A composition according to claim 13, wherein the compound of formula I is present in an amount of from 0.1% to 30% by weight, based on the weight of the organic material.

24. Use of a compound of formula I according to claim 1 as ultraviolet (UV) and visible (VIS) light absorber in coatings.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/056004

A. CLASSIFICATION OF SUBJECT MATTER
INV. C07D487/04 C07D498/04 C07D513/04 C07D471/04 C09D7/12

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C07D C09D

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

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

EPO-Internal, WPI Data, CHEM ABS Data, BEILSTEIN Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 02/28854 A (CIBA SC HOLDING AG [CH]; REINEHR DIETER [DE]; SAUTER HANSPETER [DE]; Z) 11 April 2002 (2002-04-11) page 1, paragraph 3; claims 1,18	1-10
A	EP 0 717 313 A (EASTMAN KODAK CO [US]) 19 June 1996 (1996-06-19) Formulae (Ia) and (Ib), p. 7;	1-10
A	US 6 166 218 A (RAVICHANDRAN RAMANATHAN [US] ET AL) 26 December 2000 (2000-12-26) cited in the application claim 1	1-10
A	US 5 436 349 A (WINTER ROLAND A E [US] ET AL) 25 July 1995 (1995-07-25) cited in the application claim 1	1-10

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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

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

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

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

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

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

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

"&" document member of the same patent family

Date of the actual completion of the international search 24 October 2007	Date of mailing of the international search report 31/10/2007
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Bérillon, Laurent

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2007/056004

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		US 5280124 A	18-01-1994