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METHOD FOR THE DIRECT DYEING OF HAIR AND TRICHOMAS

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(57) Abstract: The present invention relates to azomethine-type direct dyes having a triaromatic unit of formula (I) below: and also the use thereof for dyeing keratin fibres, in particular human keratin fibres such as the hair. The invention also relates to a composition for dyeing keratin fibres, comprising, in a medium suitable for dyeing, such direct dyes. Another subject of the present invention is a method for dyeing keratin fibres using said dye composition. Finally, the present invention also relates to precursors of these direct dyes.
Triaromatic azomethine direct dyes, dye composition comprising at least one such dye, method of implementation and use

The present invention relates to particular triaromatic direct dyes of azomethine type and also to the use thereof for dyeing keratin fibres, in particular human keratin fibres such as the hair.

The invention also relates to a composition for dyeing keratin fibres comprising, in a medium suitable for dyeing, such direct dyes and also to a method for dyeing using said composition.

A final subject of the invention is the precursors of these direct dyes, the use thereof for dyeing fibres and a multicomartment device containing them.

The present invention relates to the field of dyeing keratin fibres and more particularly to the field of hair dyeing.

It is known practice to dye keratin fibres, and in particular the hair, with dye compositions containing one or more direct dyes, according to a "direct dyeing" method.

The method conventionally used in direct dyeing consists in applying to the keratin fibres one or more direct dyes, or colouring molecules, which have an affinity for said fibres, leaving them on for a time, and then rinsing the fibres. The direct dyes used hitherto are generally nitrobenzene dyes, anthraquinone dyes, nitropyridine dyes, dyes ofazo, xanthene, acridine or azine type or triarylmethane-type dyes.

These direct dyes may also be applied to the keratin fibres in the presence of an oxidizing agent if it is desired to obtain a simultaneous fibre-lightening effect.

However, the colours that result therefrom are temporary or semi-permanent colours since the nature of the interactions that bind the direct dyes to the keratin fibre and their desorption from the surface and/or the core of the fibre are responsible for their weak dyeing power and their poor fastness with respect to washing, inclement weather or perspiration.

Furthermore, such direct dyes are generally sensitive to the action of oxidizing agents, which makes them difficult to use in
particular in lightening direct dye compositions that are formulated from aqueous hydrogen peroxide solution and an alkanilizing agent, which are similar to compositions used for oxidation dyeing. In other words, direct dyes are generally not very compatible with dye compositions intended to lighten the fibres and, consequently, using them in a lightening dyeing method, as an alternative to oxidation dyeing, is not yet entirely satisfactory.

These dyes also have the drawback of lacking light stability, due to the low resistance of the chromophore to photochemical attacks, which tends to lead to the colour of the keratin fibres fading over time.

Therefore there is a real need to have direct dyes that not only enable the keratin fibres to be dyed satisfactorily but that are also light-stable, capable of resulting in colours that resist the various attacks that the fibres may be subjected to, such as inclement weather, washing and perspiration, and further, that are sufficiently stable in the presence of oxidizing agents such as aqueous hydrogen peroxide solution to be able to deliver simultaneous lightening of the fibre with the advantages set out hereinabove.

These objectives are achieved with the present invention, which relates in particular to azomethine-type direct dyes having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[
\begin{align*}
\text{(I)}
\end{align*}
\]

in which formula (I):

- \( n_1 \) and \( n_2 \) represent, independently of one another, an integer equal to 0, 1, 2, 3 or 4;
- \( R \) represents:
- a linear or branched C$_1$-C$_4$ alkyl radical, optionally substituted
  with one or more identical or different radicals selected from
  hydroxyl or imidazolium, $A_{n}^-$ radicals; $A_{n}^-$ denoting a
  cosmetically acceptable anion or mixture of anions;
- a C$_1$-C$_4$ alkoxy radical;
- a halogen atom;

- $R_1$ represents:
  - a hydrogen atom;
  - a linear or branched C$_1$-$C_5$ alkyl radical, optionally substituted
    with one or more hydroxyl radicals;
  - an aminocarbonyl radical;
  - a radical of formula (II):

                   (R')$_m$

    (II)

in which formula (II):
- $m$ represents an integer equal to 0, 1, 2, 3 or 4;
- $R'$ represents:
  - a linear or branched C$_1$-C$_4$ alkyl radical, optionally
    substituted with one or more identical or different radicals
    selected from hydroxyl or imidazolium, $A_{n}^-$
    radicals; $A_{n}^-$ denoting a cosmetically acceptable anion or
    mixture of anions;
  - a C$_1$-C$_4$ alkoxy radical;
  - a halogen atom;
  - a hydroxyl radical;
  - an -NR$_3'$R'$_4$ radical in which R'$_3$ and R'$_4$ represent, independently
    of one another:
      - a hydrogen atom;
      - a linear or branched C$_1$-C$_5$ alkyl radical, optionally
        substituted with one or more hydroxyl or C$_1$-C$_4$ alkoxy
        radicals;
it being understood that, in formula (II) , when \( m \) is greater than or equal to 2, the \( R' \) radicals may be identical or different;

- \( R_2 \) represents:
  - a hydrogen atom;
  - a \( \text{C}1-\text{C}8 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;

- \( R_1 \) and \( R_2 \) may form, together with the nitrogen atom to which they are attached, a pyrrolidino, piperidino or morpholino ring;

- \( R_3 \) and \( R_4 \) represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched \( \text{C}1-\text{C}8 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - a \( \text{C}1-\text{C}4 \) alkoxy radical;

- \( X_1 \) and \( X_2 \) represent, independently of one another:
  - a hydroxyl radical;
  - an \(-\text{NR}''3 \text{R}''4\) radical in which:
    - \( R''3 \) represents:
      - a hydrogen atom;
      - a linear \( \text{C}1-\text{C}6 \) alkyl radical;
    - \( R''4 \) represents:
      - a hydrogen atom;
      - a linear or branched \( \text{C}3-\text{C}6 \) alkyl radical;
      - a linear or branched \( \text{C}2-\text{C}6 \) alkyl radical substituted with one or more hydroxyl radicals;

\( R''3 \) and \( R''4 \) may form, together with the nitrogen atom to which they are attached, a piperidino or morpholino ring;

it being understood that the compounds of formula (I) may not represent the compounds A to F below:
Another subject of the present invention relates to a composition for dyeing keratin fibres, in particular human keratin fibres such as the hair, comprising, in a medium suitable for dyeing, one or more azomethine-type dyes having a triaromatic unit of formula (I) below:

\[
\begin{align*}
\text{(I)} & \quad \text{in which formula (I):} \\
\text{• } n_1 \text{ and } n_2 \text{ represent, independently of one another, an integer equal to } 0, 1, 2, 3 \text{ or } 4; \\
\text{• } R \text{ represents:} \\
\quad & \text{- a linear or branched } C_1-C_4 \text{ alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, } A_n^- \text{ radicals; } A_n^- \text{ denoting a cosmetically acceptable anion or mixture of anions;} \\
\quad & \text{- a } C_1-C_4 \text{ alkoxy radical;} \\
\quad & \text{- a halogen atom;} \\
\text{• } R_i \text{ represents:} \\
\quad & \text{- a hydrogen atom;} \\
\quad & \text{- a linear or branched } C_i-C_s \text{ alkyl radical, optionally substituted with one or more hydroxyl radicals;} \\
\quad & \text{- an aminocarbonyl radical;} \\
\quad & \text{- a radical of formula (II):} \\
\end{align*}
\]

\[
\begin{align*}
The (II) & \quad \text{in which formula (II):} \\
\text{o } m \text{ represents an integer equal to } 0, 1, 2, 3 \text{ or } 4; \\
\text{o } R' \text{ represents:}
\end{align*}
\]
- a linear or branched C₁-C₄ alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, An⁻ radicals; An⁻ denoting a cosmetically acceptable anion or mixture of anions;
- a C₁-C₄ alkoxy radical;
- a halogen atom;
- a hydroxyl radical;
- an -NR'_₃R'_₄ radical in which R'_₃ and R'_₄ represent, independently of one another:
  ➢ a hydrogen atom;
  ➢ a linear or branched C₁-C₅ alkyl radical, optionally substituted with one or more hydroxyl or C₁-C₄ alkoxy radicals;

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10  
15  

or it being understood that, in formula (II), when m is greater than or equal to 2, the R' radicals may be identical or different;

• R₂ represents:
  - a hydrogen atom;
  - a C₁-C₅ alkyl radical, optionally substituted with one or more hydroxyl radicals;

20  

• R₁ and R₂ may form, together with the nitrogen atom to which they are attached, a pyrrolidino, piperidino or morpholino ring;

• R₃ and R₄ represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched C₁-C₅ alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - a C₁-C₄ alkoxy radical;

25  

• X₁ and X₂ represent, independently of one another:
  - a hydroxyl radical;
  - an -NR'_₃R'_₄ radical in which:
    - R'_₃ represents:
      • a hydrogen atom;

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a linear C<sub>1</sub>-C<sub>6</sub> alkyl radical;

- R’<sub>3</sub> represents:
  - a hydrogen atom;
  - a linear or branched C<sub>3</sub>-C<sub>6</sub> alkyl radical;
  - a linear or branched C<sub>2</sub>-C<sub>6</sub> alkyl radical substituted with one or more hydroxyl radicals;

R’’<sub>3</sub> and R’’<sub>4</sub> may form, together with the nitrogen atom to which they are attached, a piperidino or morpholino ring;

it being understood that the compounds of formula (I) may not represent the compounds A to D below:

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td><img src="image1.png" alt="Compound A" /></td>
<td><img src="image2.png" alt="Compound B" /></td>
</tr>
<tr>
<td><img src="image3.png" alt="Compound C" /></td>
<td><img src="image4.png" alt="Compound D" /></td>
</tr>
</tbody>
</table>

In particular, the invention also relates to the use of said dye composition for colouring keratin fibres, especially human keratin fibres such as the hair.
The present invention also relates to the use, for dyeing keratin fibres, in particular human keratin fibres such as the hair, of one or more azomethine-type dyes having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[(R)_{n_1}N\begin{array}{c}N\end{array}R_1\begin{array}{c}N\end{array}R_2\begin{array}{c}N\end{array}R_3\begin{array}{c}N\end{array}R_4(X)\begin{array}{c}X_1\end{array}\begin{array}{c}X_2\end{array}\]

(I)

in which formula (I) \(n_1, n_2, R, R_1, R_2, R_3, R_4, X_1\) and \(X_2\) have the same meanings as those indicated previously.

The invention additionally relates to a method for dyeing keratin fibres, in particular human keratin fibres such as the hair, in which applied to said fibres is a dye composition comprising, in a medium suitable for dyeing, one or more azomethine-type dyes having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[(R)_{n_1}N\begin{array}{c}N\end{array}R_1\begin{array}{c}N\end{array}R_2\begin{array}{c}N\end{array}R_3\begin{array}{c}N\end{array}R_4(X)\begin{array}{c}X_1\end{array}\begin{array}{c}X_2\end{array}\]

(I)

in which formula (I) \(n_1, n_2, R, R_1, R_2, R_3, R_4, X_1\) and \(X_2\) have the same meanings as those indicated previously; for a time sufficient to obtain the desired coloration, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.

Similarly, the invention relates more particularly to a method for lightening keratin fibres, especially human keratin fibres such as the hair, in which applied to said fibres are (i) a dye composition free
of oxidizing agent and comprising, in a medium suitable for dyeing, one or more azomethine-type dyes having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

![Chemical structure](image)

in which formula (I) \( n_1, n_2, R, R_1, R_2, R_3, R_4, X_1 \) and \( X_2 \) have the same meanings as those indicated previously; and (ii) a cosmetic composition comprising one or more oxidizing agents; compositions (i) and (ii) being applied to said keratin fibres sequentially or simultaneously for a time sufficient to obtain the desired lightening, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.

The azomethine-type direct dyes having a triaromatic unit of formula (I) according to the invention thus make it possible to result in colours that resist the various attacks that the keratin fibres may be subject to, such as inclement weather, light, washing and perspiration.

Furthermore, the direct dyes according to the invention enable the keratin fibres to be dyed satisfactorily, especially by resulting in powerful, chromatic and sparingly selective colours and lead to an improved uptake of the colouration.

The direct dyes according to the invention are light-stable and can be used in the presence of an oxidizing agent, which facilitates their use in lightening direct dye compositions based on oxidizing agents.

In other words, the direct dyes according to the present invention lead to long-lasting colours and are compatible with dye compositions that are intended to lighten keratin fibres.
Furthermore, one subject of the invention is colourless or weakly coloured leuco-type compounds, which correspond to the reduced form of the azomethine-type direct dyes having a triaromatic unit according to the invention, of formula (III) below:

![Formula III](image)

and the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof;

in which formula (III) \( n_1, n_2, R, R_1, R_2, R_3, R_4, X_i \) and \( X_2 \) have the same meanings as those indicated previously in formula (I).

The leuco-type compounds according to the invention may therefore lead, in the presence of one or more oxidizing agents, to azomethine-type direct dyes of formula (I).

Thus, the invention also relates to the use of one or more leuco-type compounds of formula (III) as precursors of the direct dyes of formula (I).

In particular, the invention relates to the use of one or more leuco-type compounds of formula (III) in the presence of one or more oxidizing agents for dyeing keratin fibres, especially human keratin fibres such as the hair.

The invention also relates to a multicompartment device or kit containing a first compartment comprising one or more leuco-type compounds of formula (III) as defined previously and a second compartment comprising one or more oxidizing agents.

The leuco-type compounds of formula (III) used under oxidizing conditions thus have the advantage of resulting in colours that resist the various attacks that the keratin fibres may be subjected to, such as inclement weather, washing, light or perspiration.
Other features, aspects, subjects and advantages of the present invention will become even more clearly apparent on reading the following description and examples.

1. Azomethine-type compound having a triaromatic unit

The direct dyes of formula (I) according to the present invention comprise in their structure at least three aromatic rings and one azomethine bond.

$A_n^-$ denotes a cosmetically acceptable anion or mixture of anions, for instance halides, such as chloride; methosulfates; nitrates; alkylsulfonates: $\text{Alk-S(0)}_2\text{O}^-$ such as methylsulfonate or mesylate and ethylsulfonate; arylsulfonates: $\text{Ar-S(0)}_2\text{O}^-$ such as benzenesulfonate and toluenesulfonate or tosylate; citrate; succinate; tartrate; lactate; alkylsulfates: $\text{Alk-S(O)}\text{SO}_2\text{O}^-$ such as methylsulfate; arylsulfates such as benzenesulfate and toluenesulfate; phosphate; acetate; triflate; and borates such as tetrafluoroborate.

Preferably, $A_n^-$ is an anionic counterion selected from bromide, chloride, methylsulfate and toluenesulfonate ions or a mixture of these ions.

According to one embodiment, in formula (I), azomethine-type direct dyes according to the invention are such that, taken together or separately:

- $n_1$ and $n_2$ represent, independently of one another, an integer equal to 0, 1 or 2;
- $R$ represents:
  - a linear or branched $\text{C}_1\text{C}_4$ alkyl radical, preferably a methyl radical;
  - a $\text{C}_1\text{C}_4$ alkoxy radical, preferably a methoxy radical;
  - a halogen atom, preferably chlorine;
- $P_i$ represents:
  - a hydrogen atom;
- a linear or branched Ci-Cs alkyl radical, optionally substituted with one or more hydroxyl radicals;
- an aminocarbonyl radical;
- a radical of formula (II):

\[
\begin{align*}
\text{(II)} & \\
R' & \quad m \\
\end{align*}
\]

in which formula (II):
- \( m \) represents an integer equal to 0, 1, 2 or 3;
- \( R' \) represents:
  - a linear or branched \( C_1-C_4 \) alkyl radical, preferably a methyl radical;
  - a \( C_1-C_4 \) alkoxy radical, preferably a methoxy radical;
  - a halogen atom, preferably chlorine;
  - a hydroxyl radical;
  - an -NR'\(_3\)R'\(_4\) radical in which \( R'3 \) and \( R'4 \) represent, independently of one another:
    \( \supseteq \) a hydrogen atom;
    \( \supseteq \) a linear or branched \( C_1-C_5 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;
- it being understood that, in formula (II), when \( m \) is greater than or equal to 2, the \( R' \) radicals may be identical or different;

- \( R_2 \) represents:
  - a hydrogen atom;
  - a linear or branched Ci-C\(_6\) alkyl radical, a butyl, methyl or ethyl radical;
- \( R_1 \) and \( R_2 \) may form, together with the nitrogen atom to which they are attached, a pyrrolidino, piperidino or morpholino ring;
- \( R_3 \) and \( R_4 \) represent, independently of one another:
  - a hydrogen atom;
- a linear or branched Ci-Cs alkyl radical, preferably a methyl or propyl radical;
- a C1-C4 alkoxy radical, preferably a methoxy radical;

• it being understood that when R1 and R2 represent a hydrogen atom then R3 and R4 cannot simultaneously denote a hydrogen atom; and/or

• X1 and X2 represent, independently of one another:
  - a hydroxyl radical;
  - an -NR3R4 radical in which:
    - R'3 represents:
      - a hydrogen atom;
      - a linear Ci-C6 alkyl radical;
    - R''4 represents:
      - a hydrogen atom;
      - a linear or branched C3-C6 alkyl radical;
      - a linear or branched C2-C6 alkyl radical substituted with one or more hydroxyl radicals;
    - R'3 and R'4 may form, together with the nitrogen atom to which they are attached, a piperidino or morpholino ring.

Preferably, n1 and n2 represent an integer equal to 0, 1 or 2, and more preferably n1 and n2 are equal to 0 or 1.

Preferably, R represents a halogen atom, in particular chlorine, or a linear C1-C4 alkyl radical, in particular a methyl radical.

Preferably, R1 represents a linear or branched C1-C4 alkyl radical, in particular a methyl, ethyl or butyl radical; a linear C1-C4 alkyl radical substituted with a hydroxyl radical, in particular a 2-hydroxyethyl radical; an aminocarbonyl radical; a radical of formula (II) in which m represents 0 or 1 and R' represents an -NR3R4 radical in which R'3 and R'4 represent a linear or branched C1-C4 alkyl radical.
More preferably, $R_i$ is selected from a methyl radical; a 2-hydroxyethyl radical; an aminocarbonyl radical; a radical of formula (II) in which $m$ represents 0 or 1 with $R'$ representing an $\text{-NR'}_3R'$ radical in which $R'_3$ and $R'_4$ represent a linear or branched $C_1$-$C_4$ alkyl radical.

In other words, $R_i$ is preferably other than a hydrogen atom.

Preferably, $R_2$ represents a hydrogen atom or a linear $C_1$-$C_4$ alkyl radical, preferably a butyl, methyl or ethyl radical.

More preferably, $R_2$ represents a hydrogen atom or a methyl radical.

More preferably still, $R_2$ represents a hydrogen atom.

Preferably, $R_3$ represents a hydrogen atom or a $C_1$-$C_4$ alkoxy radical, preferably a methoxy radical. In particular, $R_3$ represents a hydrogen atom.

Preferably, $R_4$ represents a hydrogen atom or a linear or branched $C_1$-$C_4$ alkyl radical, preferably a methyl radical. More preferably still, $R_4$ represents a hydrogen atom.

According to one embodiment, $R_3$ and $R_4$ represent a hydrogen atom.

According to one embodiment, $R_1$, $R_2$, $R_3$ and $R_4$ represent a hydrogen atom.

Preferably, $X_1$ and $X_2$ represent a hydroxyl radical or an $\text{-NR''}_3R''_4$ radical in which $R''_3$ represents a hydrogen atom or a linear $C_1$-$C_6$ alkyl radical and $R''_4$ represents a hydrogen atom or a linear or branched $C_3$-$C_6$ alkyl radical or a $C_2$-$C_6$ alkyl radical substituted with one or more hydroxyl radicals.

Preferentially, $X_1$ and $X_2$ represent a hydroxyl radical or an $\text{-NR''}_3R''_4$ radical in which $R''_3$ represents a hydrogen atom or a linear $C_1$-$C_3$ alkyl radical and $R''_4$ represents a hydrogen atom or a linear or branched $C_3$-$C_4$ alkyl radical or a $C_2$-$C_4$ alkyl radical substituted with one or more hydroxyl radicals. Preferably, $R''_3$ represents a hydrogen atom or a methyl or ethyl radical and $R''_4$ represents a hydrogen atom or an isopropyl or 2-hydroxyethyl radical.
In other words, \( X_1 \) and \( X_2 \) preferably represent a hydroxyl radical.

Alternatively, \( X_1 \) and \( X_2 \) represent an \(-N'R''_3R''_4\) radical in which \( R''_3 \) represents a hydrogen atom or a linear \( C_1-C_3 \) alkyl radical and \( R''_4 \) represents a hydrogen atom or a linear or branched \( C_3-C_4 \) alkyl radical, or a \( C_2-C_4 \) alkyl radical substituted with one or more hydroxyl radicals.

According to one embodiment, \( n_1 \) and \( n_2 \) represent an integer equal to 0 or 1, \( R_1 \) is other than a hydrogen atom, \( R_2 \) represents a hydrogen atom and \( R_4 \) represents a hydrogen atom.

According to one particular embodiment, \( n_1 \) and \( n_2 \) represent an integer equal to 0 or 1, \( R_1 \) is other than a hydrogen atom, \( R_2 \) represents a hydrogen atom, \( R_4 \) represents a hydrogen atom and \( X_1 \) and \( X_2 \) represent an \(-N'R''_3R''_4\) radical in which \( R''_3 \) represents a hydrogen atom or a linear \( C_1-C_3 \) alkyl radical and \( R''_4 \) represents a hydrogen atom or a linear or branched \( C_3-C_4 \) alkyl radical or a \( C_2-C_4 \) alkyl radical substituted with one or more hydroxyl radicals.

According to another particular embodiment, \( n_1 \) and \( n_2 \) represent an integer equal to 0 or 1, \( R_1 \) is other than a hydrogen atom, \( R_2 \) represents a hydrogen atom, \( R_4 \) represents a hydrogen atom and \( X_1 \) and \( X_2 \) represent a hydroxyl radical.

In accordance with these two particular embodiments, \( R_1 \) may preferably represent a linear or branched \( C_1-C_4 \) alkyl radical; a linear \( C_1-C_4 \) alkyl radical substituted with a hydroxyl radical; an aminocarbonyl radical; a radical of formula (II) in which \( m \) represents 0 or 1 and \( R' \) represents an \(-NR'R''_3R''_4\) radical in which \( R''_3 \) and \( R''_4 \) represent a linear or branched \( C_1-C_4 \) alkyl radical.

According to a preferred embodiment, \( X_1 \) and \( X_2 \) represent a \(-NR''_3R''_4\) radical in which \( R''_3 \) and \( R''_4 \) represent a hydrogen atom.

According to another preferred embodiment, \( X_1 \) and \( X_2 \) represent a \(-NR''_3R''_4\) radical in which \( R''_3 \) and \( R''_4 \) are different from a hydrogen atom.

According to a further embodiment, the direct dyes are chosen among the compounds of formula (I) wherein when \( X_1 \) or \( X_2 \) represent
a -NR₃R₄ radical, R₄ preferably represents a hydrogen atom or a branched or linear C₃-C₆ alkyl radical and R₃ and R₄ may form, together with the nitrogen atom to which they are attached, a piperidino ring.

According to this embodiment, R₄ does not represent a linear of branched C₂-C₆ alkyl radical substituted with one or more hydroxyl radicals and R₃ and R₄ do not form, together with the nitrogen atom to which they are attached, a morpholino ring.

Preferably, the azomethine-type direct dyes having a triaromatic unit of formula (I) according to the invention are selected from the following compound, and also the geometric or optical isomer forms thereof, the organic or inorganic acid or base salts thereof, or the solvates thereof, such as the hydrates:

<p>| Compound 1 | 2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)-phenylimino]-5-methylaminocyclohexa-2,5-dienone |
| Compound 2 | 3-Amino-6-(4-piperidinylphenylamino)-4-[4-piperidin-1-ylphenylimino]-2-propylcyclohexa-2,5-dienone |
| Compound 3 | 2-(4-Amino phenyl amino)-4-(4-amino-phenylimino)-5-(2-hydroxyethylamino)-cyclohexa-2,5-dienone |</p>
<table>
<thead>
<tr>
<th>Compound</th>
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<td>2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone</td>
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<td>5</td>
<td>5-Dibutylamino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]cyclohexa-2,5-dienone</td>
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<td>7</td>
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<td>5-Amino-2-(4-amino-2,3-dimethylphenylamino)-4-[4-amino-2,3-dimethylphenylimino]cyclohexa-2,5-dienone</td>
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<td>Compound 21</td>
<td>[4-(4-Amino-2,3-dimethylphenylamino)-6-(4-amino-2,3-dimethylphenylimino)-3-oxocyclohexa-1,4-dienyl]-urea</td>
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<td>![4-(4-Amino-2,3-dimethylphenylamino)-6-(4-amino-2,3-dimethylphenylimino)-3-oxocyclohexa-1,4-dienyl]-urea](image)</td>
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<td>{4-[4-(Ethylisopropylamino)phenylamino]-6-[4-(ethylisopropylamino)phenylimino]-3-oxocyclohexa-1,4-dienyl]urea</td>
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<td><img src="image" alt="Compound 22" /></td>
<td>![4-[4-(Ethylisopropylamino)phenylamino]-6-[4-(ethylisopropylamino)phenylimino]-3-oxocyclohexa-1,4-dienyl]urea](image)</td>
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<td>Compound 23</td>
<td>2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-dibutylaminocyclohexa-2,5-dienone</td>
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<td>54</td>
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<tr>
<td>Compound 55</td>
<td>2-(3-Chloro-4-hydroxy-phenylamino)-4-(3-chloro-4-hydroxy-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 56</td>
<td>2-(2-Chloro-4-hydroxy-phenylamino)-4-(2-chloro-4-hydroxy-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 57</td>
<td>2-(4-Amino-3(or 2)-methyl-phenylamino)-4-(4-amino-3(or 2)-methyl-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 58</td>
<td>5-Amino-2-(4-piperidin-1-ylphenylamino)-4-[4-piperidin-1-ylphenylimino]-cyclohexa-2,5-dienone</td>
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<td>67</td>
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<tr>
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2,5-Bis(4-hydroxyphenylamino)-4-(4-hydroxyphenylimino) cyclohexa-2,5-dienone

2,5-Bis(3-chloro-4-hydroxyphenylamino)-4-(3-chloro-4-hydroxyphenylimino)cyclohexa-2,5-dienone

2,5-Bis(2-chloro-4-hydroxyphenylamino)-4-(2-chloro-4-hydroxyphenylimino)cyclohexa-2,5-dienone
<p>| Compound 73 | 2,5-Bis(4-amino-3-methyl-phenylamino)-4-(4-amino-3-methylphenylimino)-cyclohexa-2,5-dienone |
| Compound 74 | 5-Amino-3-methyl-2-(4-piperidin-1-yl-phenylamino)-4-[4-piperidin-1-ylphenylimino]-cyclohexa-2,5-dienone |
| Compound 75 | 3-Amino-6-[4-(ethyl-isopropylamino)phenylamino]-4-[4-(ethyl-isopropylamino)-phenylimino]-2-propyl-cyclohexa-2,5-dienone |</p>
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<th>Compound</th>
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<td><img src="image1" alt="Compound 76" /></td>
<td>3-Amino-6-(4-amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-2-propylcyclohexa-2,5-dienone</td>
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<tr>
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<td><img src="image2" alt="Compound 77" /></td>
<td>3-Amino-6-(3-chloro-4-hydroxyphenylamino)-4-(3-chloro-4-hydroxyphenylimino)-2-propylcyclohexa-2,5-dienone</td>
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<td>78</td>
<td><img src="image3" alt="Compound 78" /></td>
<td>3-Amino-6-(2-chloro-4-hydroxyphenylamino)-4-(2-chloro-4-hydroxyphenylimino)-2-propylcyclohexa-2,5-dienone</td>
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<tr>
<td>79</td>
<td><img src="image4" alt="Compound 79" /></td>
<td>5-Amino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-3-methylcyclohexa-2,5-dienone</td>
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<td>Description</td>
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<td>80</td>
<td><img src="image" alt="Structure 80" /></td>
<td>5-Amino-2-(4-amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-3-methylcyclohexa-2,5-dienone</td>
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<tr>
<td>81</td>
<td><img src="image" alt="Structure 81" /></td>
<td>5-Amino-2-(3-chloro-4-hydroxyphenylamino)-4-(3-chloro-4-hydroxyphenylimino)-3-methylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>82</td>
<td><img src="image" alt="Structure 82" /></td>
<td>5-Amino-2-(2-chloro-4-hydroxyphenylamino)-4-(2-chloro-4-hydroxyphenylimino)-3-methylcyclohexa-2,5-dienone</td>
</tr>
</tbody>
</table>

Preferably, the direct dyes of formula (I) according to the present invention are selected from azomethine-type compounds 1, 4, 6, 16, 20, 28, 29 and 30.

The direct dyes of formula (I) may be obtained according to the procedure described below:
The azomethine-type direct dyes corresponding to formula (I) are generally obtained by reacting derivatives 1a and 1b with meta-aminophenol compounds 2 in a basic medium in the presence of an oxidizing agent. The base used is preferably an aqueous solution of ammonia or of sodium hydroxide and the oxidizing agent is preferably selected from aqueous hydrogen peroxide solution, potassium ferricyanide, air, ammonium persulfate and manganese oxide.

Methods similar to this reaction scheme are described in patent applications FR 2234277, FR 2047932, FR 2106661 and FR 2121101.

In particular, the azomethine-type direct dyes corresponding to formula (I) in which Ri corresponds to a radical of formula (II) may be obtained according to the procedure below:

The compounds corresponding to formula (Γ) are generally obtained by reacting derivatives 1 with meta-methoxyphenols 3 in a
basic medium in the presence of an oxidizing agent. The base used is preferably an aqueous solution of ammonia or of sodium hydroxide and the oxidizing agent is preferably selected from aqueous hydrogen peroxide solution, potassium ferricyanide, air, ammonium persulfate and manganese oxide.

77. Dye composition

As indicated previously, the present invention also relates to a dye composition for keratin fibres, in particular human keratin fibres such as the hair, comprising, in a medium suitable for dyeing, one or more direct dyes of formula (I) as defined previously.

The azomethine-type dyes having a triaromatic unit of formula (I) in the above composition dyeing composition preferably may not also represent the compounds E and F previously mentioned.

Preferably, the dye composition comprises one or more direct dyes of formula (I) selected from azomethine-type compounds 1, 4, 6, 16, 20, 28, 29 and 30 and also mixtures thereof.

The direct dye(s) as defined previously may be present in the dye composition according to the invention in a content ranging from 0.001% to 10% by weight, preferably in a content ranging from 0.005% to 6% by weight, relative to the total weight of the dye composition.

The dye composition according to the invention may also comprise one or more oxidation dyes.

The oxidation dyes are generally selected from oxidation bases optionally combined with one or more couplers.

By way of example, the oxidation bases are selected from \( \text{para-phenylenediamines, bis(phenyl)alkylenediamines, para-} \)

aminophenols, \( \text{ortlzo-aminophenols} \) and heterocyclic bases, and the addition salts thereof.

Among the \( \text{para-phenylenediamines, } \) mention may be made, by way of example, of \( \text{para-phenylenediamine, para-toluenediamine, 2-chloro-para-phenylenediamine, 2,3-dimethyl-para-phenylenediamine,} \)
2,6-dimethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2,6-diethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2,5-dimethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N-dimethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N-diethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N-dipropyl-\(\text{\^{\text{ara}}}-\)phenylene diamine, N,N-bis (\(\beta\)-hydroxyethyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, 4-N,N-bis (\(P\)-hydroxy-ethyl) amino -2-met hylaniline, 4-N,N-bis (\(P\)-hydroxy-ethyl) amino -2-chloro aniline, 2-\(\beta\)-hydroxyethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-fluoro-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-isopropyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N-bis (\(\beta\)-hydroxyethyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-hydroxymethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N,N,N'-dimethyl-3-methyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N-\((\beta\)-hydroxypropyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-hydroxymethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N,N,N'-dimethyl-3-methyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N,N,N'-bis (\(\beta\)-hydroxyethyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, N-(\(P\)-y-dihydroxypropyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, N-(4'-amino phenyl) -para -phenylene diamine, N,N'-bis (\(P\)-hydroxyethyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-P-hydroxyethyloxy-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-P-acetyl-aminoethyloxy-\(\text{\^{\text{ara}}}-\)phenylene diamine, N-(\(\beta\)-met hox yethyl) -para -phenylenediamine, 4-aminophenylpyrrolidine, 2-thienyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-P-hydroxyethylamino-5-aminotoluene and 3-hydroxy-1-(4'-aminophenyl)pyrrolidine, and addition salts thereof with an acid.

Among the \(\text{\^{\text{ara}}}-\)phenylenediamines mentioned above, \(\text{\^{\text{ara}}}-\)toluenediamine, 2-isopropyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-P-hydroxyethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-P-hydroxyethyloxy-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2,6-dimethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2,6-diethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2,3-dimethyl-\(\text{\^{\text{ara}}}-\)phenylenediamine, N,N,N,N'-bis (\(\beta\)-hydroxyethyl)-\(\text{\^{\text{ara}}}-\)phenylenediamine, 2-chloro -para -phenylene diamine and 2-P-acetylaminoe thyloxy-\(\text{\^{\text{ara}}}-\)phenylenediamine, and the addition salts thereof with an acid, are particularly preferred.

Among the bis(phenyl)alkylenediamines, mention may be made, by way of example, of N,N'-bis (\(P\)-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis (\(P\)-hydroxyethyl)-N,N'-bis (4'-amino phenyl) ethylene diamine, N,N'-bis(4'- amino phenyl) tetramethylene diamine, N,N'-bis (\(\beta\)-hydroxy ethyl) -N,N'-bis(4'- amino -phenyl) tetramethylene diamine, N,N'-bis(4'-met hylanimo phenyl)-
tetramethylene diamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methyl-phenyl)ethylenediamine and 1,8-bis(2,5-diaminophenoxy)-3,6-dioxaoctane, and the addition salts thereof.

Among the /?ara-aminophenols, mention may be made, by way of example, of /?ara-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-chlorophenol, 4-amino-3-(hydroxymethyl)phenol, 4-amino-2-methylphenol, 4-amino-2-(hydroxymethyl)phenol, 4-amino-2-(methoxymethyl)phenol, 4-amino-2-(aminomethyl)phenol, 4-amino-2 -[(P-hydroxyethyl)aminomethyl]phenol, 4-amino-2-fluorophenol and the addition salts thereof with an acid.

Among the o/rta-aminophenols, mention may be made, by way of example, of 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-methylphenol, 5-acetamido-2-aminophenol and the addition salts thereof.

Among the heterocyclic bases, mention may be made, by way of example, of pyridine derivatives, pyrimidine derivatives and pyrazole derivatives.

Mention may be made, among pyridine derivatives, of the compounds described, for example, in patents GB 1 026 978 and GB 1 153 196, such as 2,5-diaminopyridine, 2-(4-methoxyphenyl)-amino-3-aminopyridine, 3,4-diaminopyridine and the addition salts thereof.

Other pyridine oxidation bases of use in the present invention are the 3-aminopyrazolo[1,5-a]pyridine oxidation bases or the addition salts thereof described, for example, in patent application FR 2 801 308. Mention may be made, by way of example, of pyrazolo[1,5-a]pyrid-3-ylamine, 2-(acetylamino)pyrazolo[1,5-a]pyrid-3-ylamine, 2-(morpholin-4-yl)pyrazolo[1,5-a]pyrid-3-ylamine, 3-aminopyrazolo[1,5-a]pyridine-2-carboxylic acid, 2-methoxypyrazolo[1,5-a]pyrid-3-ylamine, (3-aminopyrazolo[1,5-a]pyrid-7-yl)methanol, 2-(3-aminopyrazolo[1,5-a]pyrid-5-yl)ethanol, 2-(3-aminopyrazolo[1,5-a]pyrid-7-yl)ethanol, (3-aminopyrazolo[1,5-a]pyrid-2-yl)methanol, 3,6-diaminopyrazolo[1,5-a]pyridine, 3,4-diaminopyrazolo[1,5-a]pyridine, pyrazolo[1,5-a]pyridine-3,7-diamine,
7-(morpholin-4-yl)pyrazolo[1,5-a]pyridine-3, 5-diamine, 5-(morpholin-4-yl)pyrazolo[1,5-a]pyridine-3-ylamine, 2-[(3-aminopyrazolo[1,5-a]pyrid-5-yl)(2-hydroxyethyl)amino]ethanol, 2-[(3-aminopyrazolo[1,5-a]pyrid-7-yl)(2-hydroxyethyl)amino]ethanol, 3-aminopyrazolo[1,5-a]pyridin-5-ol, 3-aminopyrazolo[1,5-a]pyridin-4-ol, 3-aminopyrazolo[1,5-a]pyridin-6-ol, 3-aminopyrazolo[1,5-a]pyridin-7-ol and the addition salts thereof.

Mention may be made, among pyrimidine derivatives, of the compounds described, for example, in patents DE 2359399, JP 88-169571, JP 05-63124 and EP 0 770 375 or patent application WO 96/15765, such as 2,4,5,6-tetraaminopyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triaminopyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-triaminopyrimidine and the addition salts thereof and the tautomeric forms thereof, when a tautomeric equilibrium exists.

Mention may be made, among pyrazole derivatives, of the compounds described in patents DE 3843892 and DE 4133957 and patent applications WO 94/08969, WO 94/08970, FR-A-2 733 749 and DE 195 43 988, such as 4,5-diamino-1-methylpyrazole, 4,5-diamino-1-(P-hydroxyethyl)pyrazole, 3,4-diaminopyrazole, 4,5-diamino-1-(4′-chlorobenzyl)pyrazole, 4,5-diamino-1,3-dimethylpyrazole, 4,5-diamino-3-methyl-1-phenylpyrazole, 4,5-diamino-1-methyl-3-phenylpyrazole, 4-amino-1,3-dimethyl-5-hydrazinopyrazole, 1-benzyl 1,4,5-diamino-3-methylpyrazole, 4,5-diamino-3-3-(t-butyl)-1-methylpyrazole, 4,5-diamino-1-(P-hydroxyethyl)-3-methylpyrazole, 4,5-diamino-1-(P-hydroxyethyl)-3-methylpyrazole, 4,5-diamino-1-ethyl-3-methylpyrazole, 4,5-diamino-1-ethyl-3-(4′-methoxyphenyl)pyrazole, 4,5-diamino-1-ethyl-3-(hydroxymethyl)pyrazole, 4,5-diamino-3-hydroxyethyl-1-methylpyrazole, 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole, 4,5-diamino-3-methyl-1-isopropylpyrazole, 4-amino-5-(2′-amino ethyl) amino -1,3-dimethylpyrazole, 3,4,5 -triamino pyrazole, 1-methyl 1,3,4,5 -triamino pyrazole, 3,5-diamino -1-methyl-4-(methylamino)pyrazole, 3,5-diamino -4-(P-hydroxyethyl) amino -1-
methylpyrazole and the addition salts thereof. 4,5-Diamino-1-(β-methoxyethyl)pyrazole may also be used.

Use will preferably be made of a 4,5-diaminopyrazole and more preferably still of 4,5-diamino-1-(P-hydroxyethyl)pyrazole and/or a salt thereof.

Mention may also be made, as pyrazole derivatives, of diamino-N,N-dihydropyrazolopyrazolones and in particular those described in application FR-A-2 886 136, such as the following compounds and the addition salts thereof: 2,3-diamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one, 2-amino-3-ethylamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one, 2-amino-3-isopropylamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one, 4,5-diamino-1,2-dimethyl-1,2-dihydropyrazol-3-one, 4,5-diamino-1,2-diethyl-1,2-dihydropyrazol-3-one, 4,5-diamino-1,2-dimethyl-1,2-dihydropyrazol-3-one, 4,5-diamino-1,2-diethyl-1,2-dihydropyrazol-3-one, 4,5-diamino-1,2-diethyl-1,2-dihydropyrazol-3-one, 2-amino-3-(2-hydroxyethyl)-1,2-dihydropyrazol-3-one, 2-amino-3-(2-hydroxyethyl)amino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one, 2-amino-3-dimethylamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one, 2,3-diamino-5,6,7,8-tetrahydro-1H,6H-pyridazino[1,2-a]pyrazol-1-one.

Use will preferably be made of 2,3-diamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one and/or a salt thereof.

Use will preferably be made, as heterocyclic bases, of 4,5-diamino-1-(P-hydroxyethyl)pyrazole and/or 2,3-diamino-6,7-dihydro-1H,5H-pyrazolo[1,2-a]pyrazol-1-one and/or a salt thereof.

The dye composition may optionally comprise one or more couplers advantageously selected from those conventionally used for dyeing keratin fibres.

Among these couplers, mention may especially be made of meta-phenylenediamines, meta-aminophenols, meta-diphenols,
naphthalene-based couplers and heterocyclic couplers, and also the addition salts thereof.

Mention may be made, for example, of 1,3-dihydroxybenzene, 1,3-dihydroxy-2-methylbenzene, 4-chloro-1,3-dihydroxybenzene, 2,4-diamino-1-(P-hydroxyethylxyloxy)benzene, 2-amino-4-(β-hydroxyethylamino)-1-methoxybenzene, 1,3-diaminobenzene, 1,3-bis(2,4-diaminophenoxy)propane, 3-ureidoaniline, 3-ureido-1-(dimethylamino)benzene, sesamol, 1-(P-hydroxyethylamino)-3,4-methylenedioxybenzene, cc-naphthol, 2-methyl-1-naphthol, 6-hydroxyindole, 4-hydroxyindole, 4-hydroxy-N-methylindole, 2-amino-3-hydroxypyridine, 6-hydroxybenzomorpholine, 3,5-diamino-2,6-dimethoxypyridine, 1-N-(β-hydroxyethyl)amino-3,4-methylenedioxybenzene, 2,6-bis(P-hydroxyethylamino)toluene, 6-hydroxyindoline, 2,6-dihydroxy-4-methylpyridine, 1H-3-methylpyrazol-5-one, 1-phenyl-3-methylpyrazol-5-one, 2,6-dimethylpyrazolo[1,5-b]-1,2,4-triazole, 2,6-dimethyl[3,2-c]-1,2,4-triazole, 6-methylpyrazolo[1,5-a]benzimidazole, the addition salts thereof with an acid, and mixtures thereof.

In general, the addition salts of the oxidation bases and couplers which can be used in the context of the invention are selected in particular from the addition salts with an acid, such as the hydrochlorides, hydrobromides, sulfates, citrates, succinates, tartrates, lactates, tosylates, benzenesulfonates, phosphates and acetates.

The oxidation base(s) each advantageously represent from 0.00 1% to 10% by weight and preferably from 0.005% to 6% by weight relative to the total weight of the composition.

The content of coupler(s), if it is (they are) present, each advantageously represents from 0.00 1%, to 10%> by weight relative to the total weight of the composition, and preferably from 0.005% to 6% by weight relative to the total weight of the dye composition.

The dye composition according to the invention may also comprise one or more additional direct dyes other than the azomethine-type direct dyes defined previously.
The additional direct dye(s) according to the invention are selected from neutral, acidic or cationic nitrobenzene dyes, neutral, acidic or cationic azo direct dyes, neutral, acidic or cationic quinone and in particular anthraquinone direct dyes, azine direct dyes, triarylmethane direct dyes, azomethine direct dyes and natural direct dyes.

Among the benzene direct dyes that can be used according to the invention, mention may be made, in a non-limiting manner, of the following compounds:

- 1,4-diamino-2-nitrobenzene;
- 1-amino -2-nitro -4-β-hydroxyethylamino benzene;
- 1-amino -2-nitro -4-bis (β-hydroxy ethyl) aminobenzene;
- 1,4-bis (P-hydroxyethylamino)-2-nitrobenzene;
- 1-β-hydroxy ethylamino -2-nitro -4-bis (β-
hydroxyethyl)aminobenzene;
- 1-β-hydroxy ethylamino -2-nitro -4-amino benzene;
- 1-β-hydroxy ethylamino -2-nitro -4-(ethyl 1)(β-
hydroxyethy)aminobenzene;
- 1-amino -3-methy 1-4-β-hydroxy ethylamino -6-nitrobenzene;
- 1-amino -2-nitro -4- β-hydroxy ethylamino -5-chlorobenzene;
- 1,2-diamino-4-nitrobenzene;
- 1-amino -2- β-hydroxy ethylamino -5-nitrobenzene;
- 1,2-bis^β-hydroxyethylamino)-4-nitrobenzene;
- 1-amino-2-tris(hydroxymethyl)methylamino-5-nitrobenzene;
- 1-hydroxy ethylamino -1-2- amino -5-nitrobenzene;
- 1-hydroxy ethylamino -1-2- amino -6-dinitrobenzene;
- 1-β-hydroxyethy loxy-2- β-hydroxy ethylamino -5-nitrobenzene;
- 1-met hoxy-2- β-hydroxy ethylamino -5-nitrobenzene;
- 1-β-hydroxyethoxy-3-met hylamino -4-nitrobenzene;
- 1-β,γ-dihy droxypropyloxy- 3-methy lamino -4-nitrobenzene;
- 1-P-hydroxyethylamino-4-P,Y-dihydroxypropyloxy-2-nitrobenzene;
- 1-P,Y-dihydroxypropylamino-4-trifluoromethyl-2-nitrobenzene;
- 1-P-hydroxyethylamino-4-trifluoromethyl-2-nitrobenzene;
- 1-P-hydroxyethylamino-3-methylnitrobenzene;
- 1-P-aminoethylamino-5-methoxy-2-nitrobenzene;
- 1-hydroxy-2-chloro-6-ethylamino-4-nitrobenzene;
- 1-hydroxy-2-chloro-6-amino-4-nitrobenzene;
- 1-hydroxy-6-bis-(P-hydroxyethyl)amino-3-nitrobenzene;
- 1-P-hydroxyethylamino-2-nitrobenzene; and
- 1-hydroxy-4-hydroxyethylamino-3-nitrobenzene.

Among the azo direct dyes that can be used according to the invention, mention may be made of the cationic azo dyes described in patent applications WO 95/1 5144, WO-95/0 1772 and EP-7 14954, the content of which forms an integral part of the invention.

Among these compounds, mention may be made very particularly of the following dyes:
- 1,3-dimethyl-2-[(4-(dimethylamino)phenyl)azo]-1H-imidazolium chloride,
- 1,3-dimethyl-2-[(4-aminophenyl)azo]-1H-imidazolium chloride,
- 1-methyl-4-[(methylphenylhydrazono)methyl]pyridinium methyl sulfate.

Among the azo direct dyes that may also be mentioned are the following dyes, described in the Colour Index International, 3rd edition: Disperse Red 17, Acid Yellow 9, Acid Black 1, Basic Red 22, Basic Red 76, Basic Red 51, Basic Yellow 57, Basic Brown 16, Acid Yellow 36, Acid Orange 7, Acid Red 33, Acid Red 35, Basic Brown 17, Acid Yellow 23, Acid Orange 24, Disperse Black 9.

Mention may also be made of 1-(4'-aminodiphenylazo)-2-methyl-4-bis-(P-hydroxyethyl)amino benzene and 4-hydroxy-3-(2-methoxyphenylazo)-1-naphthalenesulfonic acid.
Among the quinone direct dyes, mention may be made of the following dyes: Disperse Red 15, Solvent Violet 13, Acid Violet 43, Disperse Violet 1, Disperse Violet 4, Disperse Blue 1, Disperse Violet 8, Disperse Blue 3, Disperse Red 11, Acid Blue 62, Disperse Blue 7, Basic Blue 22, Disperse Violet 15, Basic Blue 99 and also the following compounds:

- 1-N-methylmorpholiniumpropylamino-4-hydroxyanthraquinone;
- 1-aminopropylamino-4-methylaminoanthraquinone;
- 1-aminopropylaminoanthraquinone;
- 5-β-hydroxyethyl-1,4-diaminoanthraquinone;
- 2-aminoethylaminooanthraquinone;
- 1,4-bis (P,y-dihydroxypropylamino)anthraquinone.

Among the azine dyes, mention may be made of the following compounds: Basic Blue 17 and Basic Red 2.

Among the triarylmethane dyes that can be used according to the invention, mention may be made of the following compounds: Basic Green 1, Acid Blue 9, Basic Violet 3, Basic Violet 14, Basic Blue 7, Acid Violet 49, Basic Blue 26 and Acid Blue 7.

Among the azomethine dyes that can be used according to the invention, mention may be made of the following compounds:

- 2-P-hydroxyethylamino-5-[bis(P-4'-hydroxyethyl)amino]anilino-1,4-benzoquinone;
- 2-P-hydroxyethylamino-5-(2'-methoxy-4'-amino) anilino-1,4-benzoquinone;
- 3-N-(2'-chloro-4'-hydroxy)phenylacetilamino-6-methoxy-1,4-benzoquinone;
- 3-N-(3'-chloro-4'-methylamino)phenylureido-6-methyl-1,4-benzoquinoneimine; and
- 3-[4'-N-(ethyl,carbamylmethyl)amino]phenylureido-6-methyl-1,4-benzoquinoneimine.

Among the natural direct dyes that can be used according to the invention, mention may be made of lawsone, juglone, alizarin, purpurin, carminic acid, kermesic acid, purpurogallin,
protocatechualdehyde, indigo, isatin, curcumin, spinulosin and apigenidin. Use may also be made of extracts or decoctions containing these natural dyes and in particular henna-based extracts or poultices.

Preferably, the dye composition comprises, in a medium suitable for dyeing, one or more direct dyes of formula (I) and one or more azomethine direct dyes other than the direct dyes of formula (I).

The additional direct dye(s) may be present in the dye composition in a content ranging from 0.001% to 10% by weight, preferably in a content ranging from 0.005% to 6% by weight, relative to the total weight of the composition.

The medium that is suitable for dyeing, also known as the dye support, is a cosmetic medium generally formed from water or a mixture of water and of at least one organic solvent. Examples of organic solvents that may be mentioned include C1-C4 lower alkanols, such as ethanol and isopropanol; polyols and polyol ethers, for instance 2-butoxyethanol, propylene glycol, propylene glycol monomethyl ether, diethylene glycol monoethyl ether and monomethyl ether, and also aromatic alcohols, for instance benzyl alcohol or phenoxyethanol, and mixtures thereof.

When they are present, the solvents are present in proportions preferably of between 1% and 99% by weight approximately and more preferably still of between 5% and 95% by weight approximately, relative to the total weight of the dye composition.

The dye composition may also contain various adjuvants conventionally used in hair dye compositions, such as anionic, cationic, nonionic or amphoteric surfactants or mixtures thereof, anionic, cationic, nonionic or amphoteric polymers or mixtures thereof, inorganic or organic thickeners, and in particular anionic, cationic, nonionic and amphoteric polymeric associative thickeners, antioxidants, penetrants, sequestrants, solubilizers, fragrances, buffers, dispersants, conditioning agents, for instance volatile or non-volatile, modified or unmodified silicones such as amino silicones, film-forming agents, ceramides, preserving agents and opacifiers.
The above adjuvants are generally present in an amount, for each of them, of between 0.01% and 20% by weight relative to the weight of the composition.

Needless to say, a person skilled in the art will take care to select this or these optional additional compound(s) such that the advantageous properties intrinsically associated with the dye composition in accordance with the invention are not, or are not substantially, adversely affected by the envisaged addition(s).

The pH of the dye composition in accordance with the invention is generally between 3 and 12 approximately, preferably between 5 and 11 approximately and more particularly still from 6 to 9.5. It may be adjusted to the desired value by means of acidifying or alkanizing agents customarily used in the dyeing of keratin fibres, or alternatively using standard buffer systems.

Among the acidifying agents, mention may be made, by way of example, of inorganic or organic acids, such as hydrochloric acid, orthophosphoric acid, sulfuric acid, carboxylic acids, such as acetic acid, tartaric acid, citric acid or lactic acid, or sulfonic acids.

Among the alkanizing agents, mention may be made, by way of example, of aqueous ammonia, alkali metal carbonates, alkanolamines, such as mono-, di- and triethanolamines and derivatives thereof, sodium hydroxide, potassium hydroxide and the compounds of formula (IV) below:

\[
\begin{align*}
R_a & \quad N-W-N^+R_b \\
R_c & \quad R_d
\end{align*}
\]

(IV)

in which W is a propylene residue optionally substituted with a hydroxyl group or a C1-C4 alkyl radical; R_a, R_b, R_c and R_d, which may be identical or different, represent a hydrogen atom, a C1-C4 alkyl radical or a C1-C4 hydroxyalkyl radical.

The dye composition according to the invention may be in various forms, such as in the form of liquids, creams or gels, or in any
other form that is suitable for dyeing keratin fibres, and especially human hair.

As indicated previously, the invention also relates to the use of the dye composition as defined previously for colouring keratin fibres, in particular human keratin fibres such as the hair.

III. Use of azomethine-type compounds

Likewise, the present invention consists of the use, for dyeing keratin fibres, in particular human keratin fibres such as the hair, of one or more azomethine-type direct dyes having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[
\begin{align*}
\text{(I)} & \\
\begin{array}{c}
\text{N} \\
\text{R}_1 \quad \text{N} \\
\text{R}_2 \quad \text{R}_3 \quad \text{R}_4 \\
\text{X}_1 \quad \text{X}_2
\end{array}
\end{align*}
\]

in which formula (I) \( n_1, n_2, R, R_i, R_2, R_3, R_4, X_1 \) and \( X_2 \) have the same meanings as those indicated previously.

The compounds A, B, C and D are not used for dyeing keratin fibres.

The compounds E and F may also not be used for dyeing keratin fibres.

IV. Dyeing method

The dyeing method according to the present invention consists in applying a dye composition as defined previously to the keratin fibres for a time sufficient to obtain the desired coloration, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.
The dye composition used during the keratin fibre dyeing method does not comprise the compounds A, B, C, D. Preferably, the dye composition used during the method of the present invention may also not comprise the compounds E and F.

Preferably, the leave-on time for the dye composition is between 1 and 60 minutes, preferably between 5 and 40 minutes and more preferably still between 10 and 30 minutes.

The dye composition is generally applied to the keratin fibres at ambient temperature, preferably between 25°C and 55°C.

According to one embodiment, the dye composition according to the invention is applied to the keratin fibres in the presence of one or more oxidizing agents for a time that is sufficient to obtain the desired lightening.

The oxidizing agent may be present in the dye composition or may be used separately in a cosmetic composition.

Preferably, the oxidizing agent is used separately in a cosmetic composition.

Thus, the present invention also relates to a method for lightening keratin fibres, in particular human keratin fibres such as the hair, in which applied to said fibres are (i) the dye composition as defined previously free of oxidizing agent and (ii) a cosmetic composition comprising one or more oxidizing agents; compositions (i) and (ii) being applied to said keratin fibres sequentially or simultaneously for a time sufficient to obtain the desired lightening, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.

For the purposes of the present invention, the term "sequentially" means that the oxidizing composition is applied before or after the dye composition, i.e. as a pretreatment or a post-treatment.

The oxidizing agents used are selected from hydrogen peroxide, urea peroxide, alkali metal bromates, persalts such as perborates and persulfates, peracids and oxidase enzymes (with the possible cofactors thereof), among which mention may be made of
peroxidases, 2-electron oxidoreductases such as uricases, and 4-electron oxygenases, for instance laccases.

The oxidizing agent is preferably hydrogen peroxide.

The oxidizing composition may also contain various adjuvants conventionally used in compositions for dyeing the hair and as defined previously.

The pH of the oxidizing composition containing the oxidizing agent is such that, after mixing with the dye composition, the pH of the resulting composition applied to the keratin fibres preferably ranges between 3 and 12 approximately, and more preferably still between 5 and 11 and more particularly still between 6 and 9.5. It may be adjusted to the desired value by means of acidifying or alkalinizing agents usually used in the dyeing of keratin fibres and as defined previously.

V. Leuco-type compound

Furthermore, the present invention relates to leuco-type compounds of formula (III) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[
\begin{align*}
\text{(III)} \\
\end{align*}
\]

in which formula (III) \( n_1, n_2, R, R_1, R_2, R_3, R_4, X_i \) and \( X_2 \) have the same meanings as those indicated in formula (I).

In particular, the preferred variants of \( n_1, n_2, R, R_1, R_2, R_3, R_4, X_i \) and \( X_2 \) in formula (III) for the leuco-type compounds correspond to those indicated in formula (I) for the direct dyes.

The leuco-type compounds corresponding to formula (III) are generally obtained by reacting the azomethine-type compounds of
formula (I) with a reducing agent according to the reaction scheme below:

\[
\begin{array}{c}
\text{reducing agent} \\
\end{array}
\]

Synthesis approaches similar to this reaction scheme are described in patent applications FR 2 056 799, FR 2 047 932, FR 2 165 965 and FR 2 262 023.

The leuco-type compounds of formula (III) are used as precursors of direct dyes of formula (I).

Preferably, the leuco-type compounds of formula (III) are selected from the compounds corresponding to the reduced form of the azomethine-type direct dyes 1 to 82 mentioned previously.

In other words, the leuco-type compounds of formula (III) are selected from the precursors of azomethine-type direct dyes 1 to 82.

More preferably still, the leuco-type compounds of formula (III) are selected from the precursors of direct dyes 1, 4, 6, 16, 20, 28, 29 and 30.

In particular, the invention relates to a cosmetic composition comprising one or more leuco-type compounds of formula (III) as defined previously.

The present invention also relates to a dyeing method in which a cosmetic composition comprising one or more leuco-type compounds of the abovementioned formula (III) are applied to keratin fibres in the presence of one or more oxidizing agents for a time that is sufficient to develop the desired coloration, after which the fibres are rinsed, optionally washed with shampoo, rinsed again, and the resulting fibres are dried or left to dry.

The oxidizing agent may be atmospheric oxygen or may be selected from the aforementioned oxidizing agents.
In particular, when the oxidizing agent is atmospheric oxygen, simply exposing the keratin fibres treated with the composition comprising the leuco-type compound(s) to air makes it possible to generate the colouring species and, consequently, to colour the fibres.

According to one variant, the oxidizing agent(s) may be applied to the keratin fibres simultaneously with or sequentially to the cosmetic composition comprising the leuco-type compounds.

Thus, the cosmetic composition comprising the oxidizing agent(s) may be applied to the keratin fibres before, simultaneously with or after the cosmetic composition comprising the leuco-type compounds of formula (III) according to the invention.

According to another variant, a ready-to-use composition that results from the mixing of a cosmetic composition comprising one or more leuco-type compounds of the aforementioned formula (III) and of a cosmetic composition comprising one or more oxidizing agents is applied to keratin fibres.

The ready-to-use composition which is thus applied to the keratin fibres may be in a variety of forms, such as in the form of liquids, creams or gels or in any other form appropriate for dyeing keratin fibres, and in particular human hair.

The leave-on time of the composition(s) varies from 1 to 60 minutes, preferably from 5 to 40 minutes and more preferably from 10 to 30 minutes.

The cosmetic composition comprising such leuco-type compounds is generally applied to the keratin fibres at ambient temperature, preferably between 25° C and 55° C.

Thus, the present invention also relates to a cosmetic composition, in particular for dyeing keratin fibres such as the hair, comprising one or more compounds of formula (III) as defined previously and optionally comprising one or more oxidizing agents.
vi. Dyeing device

The present invention also relates to a multicompartment device or "kit" comprising a first compartment containing a cosmetic composition comprising one or more dyes of formula (I) as defined previously or containing one or more leuco-type compounds of formula (III) as defined previously, and optionally a second compartment comprising one or more oxidizing agents.

In particular, the invention relates to a multicompartment dyeing device or kit comprising a first compartment containing a cosmetic composition comprising one or more direct dyes of formula (I) as defined previously or containing one or more leuco-type compounds of formula (III) as defined previously, and a second compartment comprising one or more oxidizing agents.

More particularly, the invention relates to a multicompartment dyeing device or kit comprising a first compartment containing a cosmetic composition comprising one or more direct dyes of the aforementioned formula (I) free of oxidizing agent and a second compartment containing a cosmetic composition comprising one or more oxidizing agents.

The invention also relates to a multicompartment dyeing device or kit comprising a first compartment containing a cosmetic composition comprising one or more leuco-type compounds of the aforementioned formula (III), and a second compartment containing a cosmetic composition comprising one or more oxidizing agents.

According to one particular embodiment, the device may comprise at least one compartment comprising a cosmetic composition comprising one or more leuco-type compounds of the aforementioned formula (III).

In this case, the composition comprising the leuco-type compound(s) as defined above is applied to the keratin fibres that are coloured by means of the exposure thereof to air.

The devices mentioned above are suitable for dyeing keratin fibres.
The evaluation of the coloration can be done visually or read on a spectrocolorimeter (such as Minolta CM3600d, illuminant D65, angle 10°, SCI values) for the L*, a*, b* colorimetric measurements. In this L*, a*, b* system, L* represents the intensity of the color, a* indicates the green/red color axis and b* indicates the blue/yellow color axis. The lower the value of L*, the darker or more intense the color is. The higher the value of a*, the redder the shade is; the higher the value of b*, the yellower the shade is.

The variation in coloring between the colored locks of natural white hair which is untreated (control) and after treatment or coloration is defined by ΔE*, corresponding to the colour uptake on keratin fibers, according to the following equation:

\[
\Delta E^* = \sqrt{\left( L^* - L_{o*} \right)^2 + \left( a^* - a_{o*} \right)^2 + \left( b^* - b_{o*} \right)^2} \tag{i}
\]

In this equation, L*, a* and b* represent the values measured after dyeing the natural hair comprising 90% of white hairs and L_{o*}, a_{o*} and b_{o*} represent the values measured for the untreated natural hair comprising 90% of white hairs.

The greater the value of ΔE*, the greater the difference in color between the control locks and the dyed locks and the greater color uptake is. Chromaticity in the CIE L*, a*, b* colorimetric system is calculated according to the following equation:

\[
C^* = \sqrt{a^{*2} + b^{*2}}
\]

The greater the value of C*, the greater the chromaticity is.

The examples that follow serve to illustrate the invention without, however, being limiting in nature.
EXAMPLES

1. Synthesis examples

Example 1: Synthesis of 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-methylamino-cyclohexa-2,5-dienone (compound 1)

Added to a solution of 0.85 g (0.004 mol) of N-(4-aminophenyl)-N-ethyl-N-isopropylamine hydrochloride in 2 ml of water and 2 ml of ethanol, is a solution of 0.40 g (0.002 mol) of 3-(methylamino)phenol hydrobromide in 1 ml of water and 2 ml of ethanol. The pH is adjusted to 9.5 with 2 ml of 20% aqueous ammonia. 7 ml of 6% aqueous hydrogen peroxide solution are added and the mixture is stirred for 24 hours. A solid is formed; after filtration, it is washed with water and then with a water/methanol mixture. The product is purified on a silica column (eluent: dichloromethane/methanol 95/5). 0.24 g of black powder of 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-methylamino-cyclohexa-2,5-dienone (compound 1) is obtained.

The molecular ion 474 (ES+) is detected by mass spectrometry.
Example 2: Synthesis of 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone (compound 4)

\[
\text{Example 2: Synthesis of 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone (compound 4)}
\]

Added to a solution of 0.42 g (0.002 mol) of N-(4-aminophenyl)-N-ethyl-N-isopropylamine hydrochloride in 2 ml of water and 2 ml of ethanol, is a solution of 0.15 g (0.001 mol) of 3-[(2-hydroxyethyl)amino]phenol. The pH is adjusted to 9.5 with 2 ml of 20% aqueous ammonia. 7 ml of 6% aqueous hydrogen peroxide solution are added and the mixture is stirred for 5 hours at ambient temperature.

The precipitate formed is filtered off, rinsed with water and dried. Thus, 80 mg of a black powder corresponding to 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone (compound 4) are obtained.

The molecular ion 504 (ES+) is detected by mass spectrometry.

Example 3: Synthesis of 5-dimethylamino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-cyclohexa-2,5-dienone (compound 6)

\[
\text{Example 3: Synthesis of 5-dimethylamino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-cyclohexa-2,5-dienone (compound 6)}
\]

Added to a solution of 0.53 g (0.003 mol) of N-(4-aminophenyl)-N-ethyl-N-isopropylamine hydrochloride in 2 ml of water and 2 ml of ethanol, is 0.41 g (0.003 mol) of 3-(dimethylamino)phenol. The pH is adjusted to 9.5 with 1.5 ml of 20% aqueous ammonia. 1 ml of
6% aqueous hydrogen peroxide solution is added and the mixture is
stirred for 48 hours. The solid formed is filtered off and then purified
by chromatography (eluent: dichloromethane/methanol 99/1).

250 mg of brown powder corresponding to 5-dimethylamino-2-
[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)-
phenylimino]cyclohexa-2,5-dienone (compound 6) are obtained.

The molecular ion 489 (ES+) is detected by mass spectrometry.

Example 4: Synthesis of 3,6-bis[4-(ethylisopropylamino) -
phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-2-methoxy-
cyclohexa-2,5-dienone (compound 28)

![Chemical Structures](attachment:image.png)

Added to a solution of 0.64 g (0.003 mol) of N-(4-
aminophenyl)-N-ethyl-N-isopropylamine hydrochloride in 2 ml of water
and 3 ml of ethanol, is a solution of 0.47 g (0.003 mol) of 2,3-
dimethoxyphenol in 1 ml of water and 3 ml of ethanol. The pH is
adjusted to 9.5 with 1.5 ml of 20% aqueous ammonia. 10.5 ml of 6%
aqueous hydrogen peroxide solution are added and the mixture is stirred
for 5 hours 30 minutes. The product is extracted with dichloromethane.
The organic phase is dried with disodium sulfate, filtered and then
concentrated until the solvents are eliminated. The product obtained is
purified by chromatography (eluent: dichloromethane/methanol 95/5).

0.12 g of black product corresponding to 3,6-bis[4-(ethyl-
isopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-
2-methoxycyclohexa-2,5-dienone (compound 28) is obtained.

The molecular ion 651 (ES+) is detected by mass spectrometry.
The H NMR spectrum is in agreement.
Example 5: Synthesis of 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-phenylaminocyclohexa-2,5-dienone (compound 29)

\[
\begin{align*}
\text{NH}_2 & \quad \text{HCl} + \quad \begin{array}{c}
\text{OH} \\
\text{NH}
\end{array} \\
\text{NH} & \quad \begin{array}{c}
\text{O}
\end{array} \\
\text{NH}
\end{align*}
\]

Compound 29

Added to a solution of 1.1 g (0.005 mol) of N-(4-aminophenyl)-N-ethyl-N-isopropylamine hydrochloride in 5 ml of water and 5 ml of ethanol, is 0.93 g (0.005 mol) of 3-hydroxydiphenylamine. The pH is adjusted to 9.5 with 20% aqueous ammonia. 17 ml of 6% aqueous hydrogen peroxide solution is added and the mixture is stirred for 48 hours. The gum formed is washed with water and then purified by chromatography (eluent: dichloromethane).

211 mg of brown powder corresponding to 2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-phenylaminocyclohexa-2,5-dienone (compound 29) are obtained.

The molecular ion 535 (ES+) is detected by mass spectrometry.

Example 6: Synthesis of 2- {4-[ethyl-(2-hydroxyethyl)amino]-2-methylphenylamino} -4- {4-[ethyl-(2-hydroxy ethyl) amino]-2-methylphenylimino} -5-phenylaminocyclohexa-2,5-dienone (compound 30)

\[
\begin{align*}
\text{NH}_2 & \quad \text{H}_2\text{SO}_4 + \quad \begin{array}{c}
\text{OH} \\
\text{NH}
\end{array} \\
\text{NH} & \quad \begin{array}{c}
\text{O}
\end{array} \\
\text{NH}
\end{align*}
\]

Compound 30
Added to a solution of 4.39 g (0.015 mol) of 2-[(4-amino-3-methylphenyl)(ethyl)amino]ethanol sulfate in 10 ml of water and 30 ml of ethanol, are 2.78 g (0.015 mol) of 3-hydroxydiphenylamine. The pH is adjusted to 9.5 with 20% aqueous ammonia. 25.5 ml of 6% aqueous hydrogen peroxide solution are added and the mixture is stirred for 24 hours. The gum formed is washed with water and then purified by chromatography (eluent: dichloromethane/methanol 9/1). 802 mg of brown powder corresponding to 2-{4-[ethyl-(2-hydroxyethyl)amino]-2-methylphenylamino} -4-{4-[ethyl-(2-hydroxyethyl)amino]-2-methylphenylimino} -5-phenylaminocyclohexa-2,5-dienone (compound 30) are obtained.

The molecular ion 568 (ES+) is detected by mass spectrometry.

Example 7:
Synthesis of 5-amino-2-[(4-amino-2,3-dimethylphenyl)amino]-4-[(4-amino-2,3-dimethylphenyl)imino]cyclohexa-2,5-dien-1-one (compound 20)

\[
\begin{align*}
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{CH}_3 & \quad \text{CH}_3 \\
\text{N} & \quad \text{N} \\
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{N} & \quad \text{N} \\
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{CH}_3 & \quad \text{CH}_3 \\
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{N} & \quad \text{O} \\
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{H}_2\text{C} & \quad \text{H}_2\text{N} \\
\text{CH}_3 & \quad \text{CH}_3 \\
\end{align*}
\]

Added to a solution of 3.14g (0.015 mol) of 2,3-dimethylbenzene-1,4-diamine and 0.817g (0.075 mol) of 3-aminophenol in 10 mL of water and 20 mL of ethanol whose the pH were adjusted to 9.5 with 20% aqueous ammonia, are 4.25 mL of 30% (0.0375 mol) aqueous hydrogen peroxide. The mixture is stirred for 24 hours. The precipitate formed is filtered off.

The solid is then dissolved in 10 mL of dimethylsulfoxide, the obtained solution is filtered and 15 mL of water is added to the filtrate.

The precipitate formed once again is filtrated and is washed with 8 mL of a mixture of dimethylsulfoxide/water (50/50) and 100 mL...
of water. The product is purified by silica chromatography (eluent dichloromethane/methanol 98/2).

804 mg of black powder corresponding to 5-amino-2-[(4-amino-2,3-dimethylphenyl)amino]-4-[(4-amino-2,3-dimethylphenyl)imino]cyclohexa-2,5-dien-1-one (compound 20) are obtained.

The molecular ion 376 (ES+) is detected by mass spectrometry.

Example 8:
Synthesis of 5-amino-2-[4-[ethyl(propan-2-yl)amino]phenyl] amino)-4-(4-[ethyl(propan-2-yl)amino]phenyl)imino) cyclohexa-2,5-dien-1-one (compound 16)

![Chemical structure]

Added to a solution of 6.44 g (0.03 mol) of N-(4-aminophenyl)-N-ethyl-N-isopropylamine and 1.65 g (0.015 mol) of 3-aminophenol in 20 mL of water and 40 mL of ethanol brought to pH 9.5 with 20% aqueous ammonia, are 15 mL of 30% aqueous hydrogen peroxide. The mixture is stirred for 10 hours. A gum is formed.

After decantation of the supernatant, the gum is washed with 20 mL of water/ethanol 50/50 mixture and the gum is added to methanol. The product crystallized for two days.

The product obtained is purified by silica chromatography (eluent dichloromethane/methanol 98/2). 1.8 g of black powder corresponding 5-amino-2-[4-[ethyl(propan-2-yl)amino]phenyl] amino)-4-(4-[ethyl(propan-2-yl)amino]phenyl)imino) cyclohexa-2,5-dien-1-one (compound 16) are obtained.

The molecular ion 460 (ES+) is detected by mass spectrometry.
II. Dyeing evaluations of the compounds synthesized

The following dye compositions were prepared:

- 500 mg of exemplified compound,
- 79 grams of water,
- 15 grams of ethanol,
- 5 grams of benzyl alcohol,
- 0.5 gram of benzoic acid.

1 gram of the mixture is applied to a 0.25 gram lock of grey hair containing 90% white hairs. After a leave-on time of 30 minutes, the lock is rinsed, washed with a standard shampoo, then rinsed again and then dried.

The results are collated in the following table:

<table>
<thead>
<tr>
<th>Compound</th>
<th>Color Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compound 1</td>
<td>Medium orange-grey</td>
</tr>
<tr>
<td>Compound 4</td>
<td>Light orange-grey</td>
</tr>
<tr>
<td>Compound 6</td>
<td>Medium orange-grey</td>
</tr>
<tr>
<td>Compound 28</td>
<td>Medium orange-grey</td>
</tr>
<tr>
<td>Compound 29</td>
<td>Medium orange-grey</td>
</tr>
<tr>
<td>Compound 30</td>
<td>Medium orange-grey</td>
</tr>
</tbody>
</table>
CLAIMS

1. Azomethine-type compound having a triaromatic unit of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

![Chemical Structure](image)

in which formula (I):

- $n_1$ and $n_2$ represent, independently of one another, an integer equal to 0, 1, 2, 3 or 4;
- $R$ represents:
  - a linear or branched C$_1$-C$_4$ alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, $A_n^-$ radicals; $A_n^-$ denoting a cosmetically acceptable anion or mixture of anions;
  - a C$_1$-C$_4$ alkoxy radical;
  - a halogen atom;
- $R_i$ represents:
  - a hydrogen atom;
  - a linear or branched C$_i$-C$_s$ alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - an aminocarbonyl radical;
  - a radical of formula (II):

![Chemical Structure](image)
in which formula (II):

\( m \) represents an integer equal to 0, 1, 2, 3 or 4;

\( R' \) represents:
- a linear or branched \( C_1 - C_4 \) alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, \( A_n' \) radicals; \( A_n' \) denoting a cosmetically acceptable anion or mixture of anions;
- a \( C_1 - C_4 \) alkoxy radical;
- a halogen atom;
- a hydroxyl radical;
- an \(-NR'_3R'_4\) radical in which \( R'_3 \) and \( R'_4 \) represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched \( C_1 - C_5 \) alkyl radical, optionally substituted with one or more hydroxyl or \( C_1 - C_4 \) alkoxy radicals;

it being understood that, in formula (II), when \( m \) is greater than or equal to 2, the \( R' \) radicals may be identical or different;

- \( R_2 \) represents:
  - a hydrogen atom;
  - a \( C_1 - C_8 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;

- \( R_i \) and \( R_2 \) may form, together with the nitrogen atom to which they are attached, a pyrrolidine, piperidine or morpholine ring;

- \( R_3 \) and \( R_4 \) represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched \( C_i - C_5 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - a \( C_1 - C_4 \) alkoxy radical;

- \( X_i \) and \( X_2 \) represent, independently of one another:
  - a hydroxyl radical;
- an -NR′′3R′′4 radical in which:
  - R′′3 represents:
    - a hydrogen atom;
    - a linear C1-C6 alkyl radical;
  - R′′4 represents:
    - a hydrogen atom;
    - a linear or branched C3-C6 alkyl radical;
    - a linear or branched C2-C6 alkyl radical substituted with
      one or more hydroxyl radicals;

R′′3 and R′′4 may form, together with the nitrogen atom to which they are attached, a piperidino or morpholino ring;

it being understood that the compounds of formula (I) may not represent the compounds A to F below:
2. Azomethine-type compound having a triaromatic unit of formula (I) according to Claim 1, characterized in that, taken together or separately:

- \( n_1 \) and \( n_2 \) represent, independently of one another, an integer equal to 0, 1 or 2;

- \( R \) represents:
  - a linear or branched \( C_1-C_4 \) alkyl radical, preferably a methyl radical;
  - a \( C_1-C_4 \) alkoxy radical, preferably a methoxy radical;
  - a halogen atom, preferably chlorine;

- \( R_1 \) represents:
  - a hydrogen atom;
  - a linear or branched \( C_1-C_8 \) alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - an aminocarbonyl radical;
  - a radical of formula (II):
in which formula (II):

- $m$ represents an integer equal to 0, 1, 2 or 3;
- $R'$ represents:
  - a linear or branched C$_1$-C$_4$ alkyl radical, preferably a methyl radical;
  - a C$_1$-C$_4$ alkoxy radical, preferably a methoxy radical;
  - a halogen atom, preferably chlorine;
  - a hydroxyl radical;
  - an $-\text{NR}_3\text{R}_4$ radical in which $\text{R}_3$ and $\text{R}_4$ represent, independently of one another:
    - a hydrogen atom;
    - a linear or branched C$_1$-C$_5$ alkyl radical, optionally substituted with one or more hydroxyl radicals;

- it being understood that, in formula (II), when $m$ is greater than or equal to 2, the $R'$ radicals may be identical or different;

- $R_2$ represents:
  - a hydrogen atom;
  - a linear or branched C$_1$-C$_6$ alkyl radical, a butyl, methyl or ethyl radical;

- $R_1$ and $R_2$ may form, together with the nitrogen atom to which they are attached, a pyrrolidine, piperidine or morpholine ring;

- $R_3$ and $R_4$ represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched C$_1$-C$_6$ alkyl radical, preferably a methyl or propyl radical;
  - a C$_1$-C$_4$ alkoxy radical, preferably a methoxy radical;

- and/or

- $X_1$ and $X_2$ represent, independently of one another:
- a hydroxyl radical;
- an \(-NR'\cdot 3R'\cdot 4\) radical in which:
  - \(R'\cdot 3\) represents:
    - a hydrogen atom;
    - a linear \(C_1-C_6\) alkyl radical;
  - \(R'\cdot 4\) represents:
    - a hydrogen atom;
    - a linear or branched \(C_3-C_6\) alkyl radical;
    - a linear or branched \(C_2-C_6\) alkyl radical substituted with one or more hydroxyl radicals.

3. Azomethine-type compound having a triaromatic unit of formula (I) according to Claim 1 or 2, characterized in that \(n_1\) and \(n_2\) represent an integer equal to 0 or 1, \(R_1\) is other than a hydrogen atom, \(R_2\) represents a hydrogen atom and \(R_4\) represents a hydrogen atom.

4. Azomethine-type compound having a triaromatic unit of formula (I) according to any one of the preceding claims, \(n_1\) and \(n_2\) represent an integer equal to 0 or 1, \(R_1\) is other than a hydrogen atom, \(R_2\) represents a hydrogen atom, \(R_4\) represents a hydrogen atom and \(X_1\) and \(X_2\) represent an \(-N'\cdot R'\cdot 3R'\cdot 4\) radical in which \(R'\cdot 3\) represents a hydrogen atom or a linear \(C_1-C_3\) alkyl radical and \(R'\cdot 4\) represents a hydrogen atom or a linear or branched \(C_3-C_4\) alkyl radical or a \(C_2-C_4\) alkyl radical substituted with one or more hydroxyl radicals.

5. Azomethine-type compound having a triaromatic unit of formula (I) according to any one of the preceding claims, characterized in that \(X_1\) and \(X_2\) represent a \(-NR'\cdot 3R'\cdot 4\) radical wherein \(R'\cdot 3\) and \(R'\cdot 4\) represent hydrogen atom.

6. Azomethine-type compound a triaromatic unit of formula (I) according to any one of the preceding claims, characterized in that \(X_1\) and \(X_2\) represent a \(-NR'\cdot 3R'\cdot 4\) radical wherein \(R'\cdot 3\) and \(R'\cdot 4\) represent hydrogen atom.
7. Azomethine-type compound a triaromatic unit of formula (I) according to any one of claims 1 to 5, characterized in that Xi and X₂ represent a -NR‴₃R‴₄ radical wherein R‴₃ and R‴₄ are different from hydrogen atom.

8. Azomethine-type compound having a triaromatic unit of formula (I) according to any one of the preceding claims, characterized in that it is selected from the following compounds and also the geometric or optical isomer forms thereof, the organic or inorganic acid or base salts thereof, or the solvates thereof, such as the hydrates:

<p>| Compound 1 | 2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)-phenylimino]-5-methylaminocyclohexa-2,5-dienone |
| Compound 2 | 3-Amino-6-(4-piperidin-1-ylphenylamino)-4-[4-piperidin-1-ylphenylimino]-2-propylcyclohexa-2,5-dienone |
| Compound 3 | 2-(4-Aminophenyl amino)-4-(4-amino-phenylimino)-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone |
| Compound 4 | 2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone |
| Compound 5 | 5-Dibutylamino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino)cyclohexa-2,5-dienone |
| Compound 6 | 5-Dimethylamino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethyl-isopropylamino)phenylimino)cyclohexa-2,5-dienone |</p>
<table>
<thead>
<tr>
<th>Compound</th>
<th>Molecular Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compound 7</td>
<td>2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-pyrrolidin-1-ylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 8</td>
<td>2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-piperidin-1-ylcyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 9</td>
<td>2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-morpholin-4-ylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 10</td>
<td>2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-methylaminocyclohexa-2,5-dienone</td>
</tr>
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<td>Compound</td>
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<tr>
<td>Compound 14</td>
<td><img src="image" alt="Compound 14" /></td>
</tr>
<tr>
<td>Compound 15</td>
<td>2-(4-Amino-3(or 2)-methylphenylamino)-4-(4-amino-3-methylphenylimino)-5-methylamino-cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>-------------</td>
<td>---------------------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Compound 16</td>
<td>5-Amino-2-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 17</td>
<td>2-(4-Hydroxyphenylamino)-4-(4-hydroxyphenylimino)-5-methylaminocyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 18</td>
<td>2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-(2-hydroxy ethylamino) - cyclohexa-2,5-dienone</td>
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<td>Compound</td>
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<tr>
<td>Compound 24</td>
<td>2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-dimethylaminocyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 25</td>
<td>2-(4-Aminophenylamino)-4-(3-chloro-4-hydroxyphenylimino)-5-(2-hydroxyethylamino)cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 26</td>
<td>4-(3-Chloro-4-hydroxyphenylimino)-2-(4-hydroxyphenylamino)-5-methylaminocyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 27</td>
<td>2,5-Bis(4-hydroxyphenylamino)-4-(4-hydroxyphenylimino)cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound</td>
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<tr>
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<tr>
<td>Compound 39</td>
<td>3,6-Bis(4-amino-3-methylphenylamino)-4-(4-amino-3-methylphenylimino)-2-methoxycyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>-------------</td>
<td>--------------------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Compound 40</td>
<td>3,6-Bis(4-hydroxyphenylamino)-4-(4-hydroxyphenylimino)-2-methoxycyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 41</td>
<td>5-Amino-2-(2-chloro-4-hydroxyphenylamino)-4-[2-chloro-4-hydroxyphenylimino]cyclohexa-2,5-dienone</td>
</tr>
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3,6-Bis(4-amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-2-methoxycyclohexa-2,5-dienone

2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-o-tolylaminocyclohexa-2,5-dienone

2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-o-tolylaminocyclohexa-2,5-dienone
Compound 45

2-(4-Aminophenylamino)-4-(4-amino-phenylimino)-5-o-tolylaminocyclohexa-2,5-dienone

Compound 46

2-(4-Hydroxy-phenylamino)-4-(4-hydroxyphenylimino)-5-o-tolylaminocyclohexa-2,5-dienone

Compound 47

2-(3-Chloro-4-hydroxy-phenylamino)-4-(3-chloro-4-hydroxy-phenylimino)-5-o-tolylaminocyclohexa-2,5-dienone

Compound 48

2-(2-Chloro-4-hydroxy-phenylamino)-4-(2-chloro-4-hydroxy-phenylimino)-5-o-tolylaminocyclohexa-2,5-dienone
<table>
<thead>
<tr>
<th>Compound</th>
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<tbody>
<tr>
<td>49</td>
<td><img src="image1.png" alt="Image" /></td>
<td>2-(4-Amino-3(or 2)-methylphenylamino)-4-(4-amino-3(or 2)-methylphenylimino)-5-o-tolylaminocyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>50</td>
<td><img src="image2.png" alt="Image" /></td>
<td>5-Amino-2-(4-morpholin-4-ylphenylamino)-4-[4-morpholin-4-ylphenylimino]cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>51</td>
<td><img src="image3.png" alt="Image" /></td>
<td>2-[4-(Ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-5-p-tolylaminocyclohexa-2,5-dienone</td>
</tr>
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<td>Chemical Structure</td>
<td>Description</td>
</tr>
<tr>
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</tr>
<tr>
<td>52</td>
<td><img src="image" alt="Compound 52" /></td>
<td>2-(4-Amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
</tr>
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<td>53</td>
<td><img src="image" alt="Compound 53" /></td>
<td>2-(4-Aminophenyl amino)-4-(4-amino-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<td>54</td>
<td><img src="image" alt="Compound 54" /></td>
<td>2-(4-Hydroxyphenylamino)-4-(4-hydroxyphenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<td>Chemical Formula</td>
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<tr>
<td>55</td>
<td><img src="image" alt="Structure 55" /></td>
<td>2-(3-Chloro-4-hydroxy-phenylamino)-4-(3-chloro-4-hydroxy-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<tr>
<td>56</td>
<td><img src="image" alt="Structure 56" /></td>
<td>2-(2-Chloro-4-hydroxy-phenylamino)-4-(2-chloro-4-hydroxy-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<td>57</td>
<td><img src="image" alt="Structure 57" /></td>
<td>2-(4-Amino-3(or 2)-methyl-phenylamino)-4-(4-amino-3(or 2)-methyl-phenylimino)-5-p-tolylaminocyclohexa-2,5-dienone</td>
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<tr>
<td>58</td>
<td><img src="image" alt="Structure 58" /></td>
<td>5-Amino-2-(4-piperidin-1-ylphenylamino)-4-[4-piperidin-1-ylphenylimino]-cyclohexa-2,5-dienone</td>
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<tr>
<td>Compound</td>
<td>Chemical Structure</td>
<td>Chemical Formula</td>
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<tr>
<td>59</td>
<td><img src="image" alt="Compound 59" /></td>
<td>2,5-Bis[4-(ethylisopropylamino)-phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-3-methylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>60</td>
<td><img src="image" alt="Compound 60" /></td>
<td>2,5-Bis(4-amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)-3-methylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>61</td>
<td><img src="image" alt="Compound 61" /></td>
<td>2,5-Bis(4-aminophenylamino)-4-(4-aminophenylimino)-3-methylcyclohexa-2,5-dienone</td>
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<tr>
<td>62</td>
<td><img src="image" alt="Compound 62" /></td>
<td>2,5-Bis(4-hydroxyphenylamino)-4-(4-hydroxyphenylimino)-3-methylcyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 63</td>
<td>2,5-Bis(3-chloro-4-hydroxyphenylamino)-4-(3-chloro-4-hydroxyphenylimino)-3-methylcyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 64</td>
<td>2,5-Bis(2-chloro-4-hydroxyphenylamino)-4-(2-chloro-4-hydroxyphenylimino)-3-methylcyclohexa-2,5-dienone</td>
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<tr>
<td>Compound 65</td>
<td>2,5-Bis(4-amino-3(or 2)-methylphenylamino)-4-(4-amino-3(or 2)-methylphenylimino)-3-methylcyclohexa-2,5-dienone</td>
<td></td>
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<tr>
<td>Compound 66</td>
<td>5-Amino-3-methyl-2-(4-morpholin-4-ylphenylamino)-4-[4-morpholin-4-ylphenylimino]cyclohexa-2,5-dienone</td>
<td></td>
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<tr>
<td>Compound</td>
<td>Chemical Structure</td>
<td></td>
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<tr>
<td>67</td>
<td><img src="image1.png" alt="Image of Compound 67" /></td>
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</tr>
<tr>
<td>68</td>
<td><img src="image2.png" alt="Image of Compound 68" /></td>
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<tr>
<td>69</td>
<td><img src="image3.png" alt="Image of Compound 69" /></td>
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<tr>
<th>Compound</th>
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<tbody>
<tr>
<td>67</td>
<td>2,5-Bis[4-(ethylisopropylamino)-phenylamino]-4-[4-(ethylisopropylamino)-phenylimino]cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>68</td>
<td>2,5-Bis(4-amino-2,3-dimethylphenylamino)-4-(4-amino-2,3-dimethylphenylimino)cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>69</td>
<td>2,5-Bis(4-amino-phenylamino)-4-(4-aminophenylimino)cyclohexa-2,5-dienone</td>
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<tr>
<td>Compound</td>
<td>Chemical Structure</td>
</tr>
<tr>
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<tr>
<td>70</td>
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<td>71</td>
<td><img src="image" alt="Compound 71" /></td>
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<tr>
<td>72</td>
<td><img src="image" alt="Compound 72" /></td>
</tr>
<tr>
<td>Compound 73</td>
<td>2,5-Bis(4-amino-3-methylphenylamino)-4-(4-amino-3-methylphenylimino)-cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 74</td>
<td>5-Amino-3-methyl-2-(4-piperidin-1-ylphenylamino)-4-[4-piperidin-1-ylphenylimino]cyclohexa-2,5-dienone</td>
</tr>
<tr>
<td>Compound 75</td>
<td>3-Amino-6-[4-(ethylisopropylamino)phenylamino]-4-[4-(ethylisopropylamino)phenylimino]-2-propylcyclohexa-2,5-dienone</td>
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<tr>
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<tr>
<td>76</td>
<td><img src="image" alt="Compound 76" /></td>
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<td>77</td>
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<tr>
<td>78</td>
<td><img src="image" alt="Compound 78" /></td>
</tr>
<tr>
<td>79</td>
<td><img src="image" alt="Compound 79" /></td>
</tr>
</tbody>
</table>
9. Composition, in particular dye composition, for keratin fibres such as the hair, characterized in that it comprises, in a medium suitable for dyeing, one or more compounds of azomethine type having a triaromatic unit of formula (I) below and also the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

![Chemical Structure](image)

\[ (I) \]
in which formula (I):
  - \(n_1\) and \(n_2\) represent, independently of one another, an integer equal to 0, 1, 2, 3 or 4;
  - \(R\) represents:
    - a linear or branched \(C_1\)-\(C_4\) alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, \(A_n^-\) radicals; \(A_n^-\) denoting a cosmetically acceptable anion or mixture of anions;
    - a \(C_1\)-\(C_4\) alkoxy radical;
    - a halogen atom;
  - \(R_i\) represents:
    - a hydrogen atom;
    - a linear or branched \(C_i\)-\(C_s\) alkyl radical, optionally substituted with one or more hydroxyl radicals;
    - an aminocarbonyl radical;
    - a radical of formula (II):

\[
\text{(II)}
\]

in which formula (II):
  - \(m\) represents an integer equal to 0, 1, 2, 3 or 4;
  - \(R'\) represents:
    - a linear or branched \(C_1\)-\(C_4\) alkyl radical, optionally substituted with one or more identical or different radicals selected from hydroxyl or imidazolium, \(A_n^-\) radicals; \(A_n^-\) denoting a cosmetically acceptable anion or mixture of anions;
    - a \(C_1\)-\(C_4\) alkoxy radical;
    - a halogen atom;
    - a hydroxyl radical;
    - an \(-NR'_3R'_4\) radical in which \(R'_3\) and \(R'_4\) represent, independently of one another:
      - a hydrogen atom;
- a linear or branched C1-C5 alkyl radical, optionally substituted with one or more hydroxyl or C1-C4 alkoxy radicals;

- it being understood that, in formula (II), when m is greater than or equal to 2, the R' radicals may be identical or different;

- R2 represents:
  - a hydrogen atom;
  - a C1-C8 alkyl radical, optionally substituted with one or more hydroxyl radicals;

- Ri and R2 may form, together with the nitrogen atom to which they are attached, a pyrrolidine, piperidine or morpholine ring;

- R3 and R4 represent, independently of one another:
  - a hydrogen atom;
  - a linear or branched C1-C8 alkyl radical, optionally substituted with one or more hydroxyl radicals;
  - a C1-C4 alkoxy radical;

- Xi and X2 represent, independently of one another:
  - a hydroxyl radical;
  - an -NR''R'''4 radical in which:
    - R''3 represents:
      - a hydrogen atom;
      - a linear C1-C6 alkyl radical;
    
    - R''4 represents:
      - a hydrogen atom;
      - a linear or branched C3-C6 alkyl radical;
      - a linear or branched C2-C6 alkyl radical substituted with one or more hydroxyl radicals;

- R''3 and R'''4 may form, together with the nitrogen atom to which they are attached, a piperidino or morpholino ring;
it being understood that the compounds of formula (I) may not represent the compounds A to D below:

<table>
<thead>
<tr>
<th>Compound A</th>
<th>Compound B</th>
</tr>
</thead>
<tbody>
<tr>
<td><img src="image1" alt="Compound A" /></td>
<td><img src="image2" alt="Compound B" /></td>
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<tr>
<td>Compound C</td>
<td>Compound D</td>
</tr>
<tr>
<td><img src="image3" alt="Compound C" /></td>
<td><img src="image4" alt="Compound D" /></td>
</tr>
</tbody>
</table>

10. Use, for dyeing keratin fibres, in particular human keratin fibres such as the hair, of one or more direct dyes of formula (I) below and also the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

![Formula](image5)

in which formula (I) \(n_1, n_2, R, R_i, R_2, R_3, R_4, X_1\) and \(X_2\) have the same meanings as those indicated in Claim 9.

11. Method for dyeing keratin fibres, in particular human keratin fibres such as the hair, in which applied to said fibres is a dye composition comprising, in a medium suitable for dyeing, one or more
direct dyes of formula (I) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[
\begin{align*}
\text{(I)} & \\
& \begin{array}{c}
\text{R}_1 \\
\text{R}_2 \\
\text{R}_3 \\
\text{R}_4 \\
\text{X}_1 \\
\text{X}_2 \\
\text{N} \\
\text{N} \\
\text{O} \\
\text{N} \\
\text{N} \\
\text{H} \\
\text{H} \\
\text{N} \\
\text{N} \\
\text{R}_{n1} \\
\text{R}_{n2}
\end{array}
\end{align*}
\]

where \( n_1, n_2, R, R_i, R_2, R_3, R_4, X_1 \) and \( X_2 \) have the same meanings as those indicated in Claim 9; for a time sufficient to obtain the desired coloration, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.

12. Method for lightening keratin fibres, in particular human keratin fibres such as the hair, in which applied to said fibres are (i) the dye composition as defined according to Claim 9 free of oxidizing agent and (ii) a cosmetic composition comprising one or more oxidizing agents; compositions (i) and (ii) being applied to said keratin fibres sequentially or simultaneously for a time sufficient to obtain the desired lightening, after which the fibres are rinsed, optionally washed with shampoo, rinsed again and the resulting fibres are dried or left to dry.

13. Leuco-type compound of formula (III) below, the organic or inorganic acid or base salts thereof, the tautomeric, optical isomer or geometric isomer forms thereof and/or the solvates thereof:

\[
\begin{align*}
\text{(III)} & \\
& \begin{array}{c}
\text{R}_1 \\
\text{R}_2 \\
\text{R}_3 \\
\text{OH} \\
\text{H} \\
\text{N} \\
\text{N} \\
\text{R}_{n1} \\
\text{R}_{n2}
\end{array}
\end{align*}
\]
in which formula (III) \( n_1, n_2, R, R_i, R_2, R_3, R_4, X_i \) and \( X_2 \) have the same meanings as those indicated in Claim 9.

14. Composition, in particular for dyeing keratin fibres such as the hair, comprising one or more compounds of formula (III) as defined according to the preceding claim and optionally comprising one or more oxidizing agents.

15. Method for dyeing keratin fibres, characterized in that a dye composition comprising one or more compounds of formula (III) as defined according to Claim 13 and an oxidizing composition comprising one or more oxidizing agents are applied simultaneously or sequentially to said wet or dry fibres.

15. Use of one or more leuco-type compounds of formula (III) as defined according to Claim 13 as precursors of the direct dyes as defined according to any one of Claims 1 to 6.

16. Use of one or more leuco-type compounds of formula (III) as defined according to Claim 13 in the presence of one or more oxidizing agents for dyeing keratin fibres, in particular human keratin fibres such as the hair.

17. Multicompartment device or "kit" comprising a first compartment containing a cosmetic composition comprising one or more dyes of formula (I) as defined in Claim 9 or containing one or more leuco-type compounds of formula (III) as defined according to Claim 13, and a second compartment comprising one or more oxidizing agents.
INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

INV. C09B55/00 A61K8/34 A61K8/35 A61K8/41 A61K8/42
A61Q5/06 A61Q5/10

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols):
C09B A61K A61Q

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 practicable, search terms used):
EPO-Internal, WPI Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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<td>JP 8 259509 A (NI PPON KANKO SHI KISO KENKYUSHO) 8 October 1996 (1996-10-08) abstract page 4; compound 6</td>
<td>1-5, 7</td>
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<tr>
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</tbody>
</table>

Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:
  - A: document defining the general state of the art which is not considered to be of particular relevance
  - E: earlier application or patent but published on or after the international filing date
  - L: document which may throw doubts on priority claim(s) or which is cited to establish the publication date of 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

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*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

*Z* document member of the same patent family

Date of the actual completion of the international search 21 March 2013

Date of mailing of the international search report 08/04/2013

Name and mailing address of the ISA:
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016

Authorized officer: Durand-Oral, 11 knur

Form PCT/ISA/210 (second sheet) (April 2005)
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