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(54) **Title:** HAIR COLOURING COMPOSITION USING PLANT DYES

(57) **Abstract:** Plant dye based hair colour composition/ kit for dyeing/ colouring the hair free of any pH controlling agent and oxidizing agent comprising selectively at least one or more dye, plant dye, concentrated plant dye and at least one or more inorganic metal salt based mordanting agent for generating dye/concentrated plant dye metal complex free of any pH controlling agents and oxidizing agents, which while providing for safe base colour shade of the dye, plant dye, concentrated plant dye also provides for safe modified base colour shade thereby displaying superior colour delivery and colour variance.

TITLE: HAIR COLOURING COMPOSITION USING PLANT DYES**FIELD OF INVENTION**

The present invention relates to dyes/ plant dyes for dyeing keratin fibres, and more particularly, relates to said plant dye based hair colour composition/ kit for dyeing/ colouring the hair free of any pH controlling agent and oxidizing agent. Particularly, the said plant dye based hair colour composition/ kit of the present invention comprises selectively at least one or more dye, plant dye, concentrated plant dye and at least one or more inorganic metal salt based mordanting agent for generating dye/concentrated plant dye metal complex free of any pH controlling agents and oxidizing agents, which while providing for safe base colour shade of the dye, plant dye, concentrated plant dye also provides for safe modified base colour shade thereby displaying superior colour delivery and colour variance.

Advantageously, the invention provides for a hair colour composition/ kit for dyeing keratinous fibers comprising selectively at least one dye, plant dye, concentrated plant dye and at least one or more inorganic metal salt based mordanting agent for generating dye/ plant dye/concentrated plant dye-metal complex free of any pH controlling agents and oxidizing agents, with safe modified base colour and/ or base colour of the dye/ plant dye/ concentrated plant dye. Advantageously, the invention favours achieving the much desired variance of the base colour of dye/ plant dye/ concentrated plant dye when the same is present in combination with different metal mordants for the purposes of dyeing of keratin fibres involving a much simple and cost-effective route free of any pre-mordanting and post-mordanting steps under ambient temperature conditions and in the process providing much desired broadening of the range of colours for such purposes that may be generated out of plant dyes suitable of such dyeing purposes.

BACKGROUND ART

Metal complex dyes are pre-metallised dyes, in which one or two dye molecules are coordinated with a metal ion. Mordanting of dyes in this way can work in a way wherein the positively charged metal ions are capable of interacting quite strongly with the hair. This is especially true for copper and more so in damaged hair which has a higher negative charge than undamaged hair. The metal ions can then interact with the (i.e. form complexes with) the aromatic plant hair dye molecules, thus locking them to the hair. Whether this effect occurs at the hair surface or within the hair depends to some extent on the size of the dye

molecule and the strength of the hair/metal ion/dye interaction. The stronger the interaction the more likely the dye complex is to be held close to the surface of the hair.

If the metal ion/dye complex is particularly strong then a water insoluble complex is formed to be fixed just prior to use. The coloured insoluble complex is then precipitated on and around the surface of the hair.

Several natural hair colouring compositions are known in the art for dyeing keratin fibers involving natural hair colouring dyes along with mordanting agents.

10 **US: 7550014 and WO 2007130777 by Greaves et al.** teaches a composition for dyeing keratin fibres, comprising approximately 0.1 to 30 percent of at least one concentrated plant dye extracted using at least one of a solvent extraction process and a supercritical CO₂ extraction process, and a mineral or metallic salt with approximately 0.01 to 5 percent active metal capable of acting as a mordanting agent, and wherein at least one of the concentrated plant dye and the mineral or metal salt capable of acting as a mordanting agent is encapsulated in a water impermeable shell. The crux of this prior art is mainly based on the attainment of substantially pure plant dyes by solvent or supercritical CO₂ extraction, and mordanting with mineral or metallic salts in combination with or without encapsulation, to create safe hair dyes that unexpectedly and surprisingly produce bright and permanent or semi-permanent colours similar to synthetic dyes in 10-20 minutes or less, thereby improving on the safety, durability, quality and application time of existing products on the market.

20 The said dyes and mineral salts are packaged separately in aqueous solutions and are mixed together just before use. The metals act as a mordant which helps to hold the dyes on the hair to provide for a permanent colouring effect against shampooing. The colour solution contains citric acid and lactic acid whereby the final colouring solution is at quite low pH values.

25 **US2009/0249563 also by Greaves et al.** teaches a composition for dyeing keratin fibers, comprising: approximately 0.1 to 30 percent of at least one concentrated plant dye; and a mineral or metallic salt with approximately 0.01 to 5 percent active metal capable of acting as a mordanting agent, wherein the mineral or metallic salt is selected from the group consisting of iron gluconate, ferrous aspartate, copper gluconate, calcium gluconate, calcium aspartate, sodium gluconate, magnesium gluconate, magnesium aspartate,

magnesium citrate, magnesium palmitate, zinc gluconate, zinc aspartate, and manganese gluconate, which are essentially organic acid based mordanting agents.

US4801302 by Grollier et al. discloses a process for dyeing hair in several separate steps consisting in applying a cosmetically acceptable composition containing at least one cupric salt and, before or after this application, a cosmetically acceptable composition containing a dye chosen from brazilin and its hydroxyl derivative wherein the copper content in the composition containing said cupric salt is between 0.01 and 2% by weight, and said dye is present in the composition containing it in a proportion between 0.05 and 5% by weight.

The cupric salt is selected from Cupric- chloride, sulphate, nitrate, acetate were used in combination with Haematoxylin dye and/ or Brazilin dye to give different colour shades under different pH conditions adjusted by external source of alkali/ acids.

US4004877 by Saphir et al teaches about aerial oxidation of natural and synthetic hair comprising of an air oxidation hair dye, a metal complex compound and a solvent which keeps the air oxidation hair dye and the metal complex compound in solution, said metal complex compound consisting of a metal present in a quantity ranging from 0.1% to less than 1% by weight and selected from the group consisting of copper, iron, manganese, cobalt, nickel, chromium, titanium, tin, hafnium, zinc, vanadium, zirconium and molybdenum; a complex forming agent present in the weight percent range corresponding to that of said metal quantity given above, and monoethanolamine ranging from 1.0% to 1.36% by weight, said solvent ranging from 38.3% to 55.25% by weight, said solvent comprising formamide said percent by weight referring to the weight of the total composition. The copper metal in the said air oxidation hair dye is present in the form of copper sulphate, said complex forming agent is tartaric acid, and wherein said solvent includes isopropyl alcohol and formamide. The aforesaid cited art involves an amine and a complexing agent such as tartaric acid in its air oxidation hair dye formulation.

US20030066140 by Bartolone et al.: is directed to a composition for coloring hair which comprises a first composition comprising: (a) a dye forming transition metal salt or complex; and a second composition which comprises the following two compositions which are mixed just prior to application to the hair: (a) a composition comprising a water-soluble peroxygen oxidizing agent; and (b) a composition comprising one or more oxidative hair coloring agents selected from the group consisting of an aromatic diamine, an aminophenol,

a polyhydric phenol a catechol and mixtures thereof. The aforesaid prior art involves the use of a water-soluble peroxygen oxidizing agent in its hair colouring composition

Regarding "**Natural hair dyeing agent prepared by plant pigment catechu and hair dyeing method by Hou et al.**", the same teaches a hair dyeing agent comprising two parts, agent A and agent B. Agent A comprises with: 1. (by wt%) 3-10% of plant color matter catechu, a traditional Chinese medicinal material. 2. 10-50% of organic solvent. 3. Water Agent B comprises with: 1. (by wt %) 1-5% of ferrous salt mordant. 2. 0.2-1.0% of reduced iron powder. 3. 1-10% of surfactant. 4. 1-5% of thickening agent. 5. Acidifying agent. 6. 0.1-0.3% of natural essence. 7. Water This prior art involves acidifying agent in its composition.

US5725602 by Balcour Carbo et al., is directed to a hair dyeing process comprising the steps of: a) obtaining a composition in the form of an aqueous dispersion containing a product of grinding plants or parts of plants of the species *Impatiens balsamina*, in an appropriate liquid carrier, said dispersion forming a pulp which has a consistency that is sufficient to coat the hair and to adhere thereto without running after the application to the hair; b) bringing the hair to be dyed into contact with the composition as obtained in a) above; c) and maintaining the said contact for a sufficient period to obtain the desired colour. The above prior art does not involve any mordanting agent.

Regarding "**A novel permanent acid type hair color mode possible with dye metal ion technology**" in *J. Cosmetic. Sci. 56: 29-46, 2005* by **Ochiai et al**, the same is based on the interaction of various metal ions namely $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, ZnCl_2 , $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$, $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, NaCl , KCl , $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$, in permanent hair dyes. Specifically the superior efficacy of $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ (optimum concentration 0-1.6%) in dye uptake is disclosed and the contribution of the acidic compounds such as formic acid, lactic acid, acetic acid, glycine hydrochloride, tretronic acid and specifically glycolic acid (concentration 0-3.2%) in color brightness and in long retention of the color is also taught. The results revealed that glycolic acid can provide colour brightness and longevity effects when compared to other acids. Further the author made an attempt to find out the optimum concentration of glycolic acids. Concentrations ranges from 0 to 3.2 % of glycolic acid were investigated and the colour brightness and longevity effects were optimum at 1.6 %. The study clearly indicated that further increment of glycolic acid concentration does not improve the colour brightness and colour longevity behaviour.

Therefore the above prior art involves only one type of counter anion for each of the cations and further involves acidic compounds for dyeing the hair.

US6849096 by Taguchi et al., teaches a hair-dyeing method comprising the steps of: first
5 mordanting hair with a dye mordant selected from the group consisting of a metal element of aluminum, iron, zinc, nickel, calcium, or magnesium, a metal salt thereof, and a mixture thereof; then secondly uniformly applying a hair dye comprising a formulation primarily composed of a dye component including an indican derived from an indigo plant and a formulation primarily composed of β -glucosidase enzyme component, the hair dye being
10 applied to hair either directly or after mixed with a proper amount of water; and, upon completion of hair dyeing, finishing the hair by rinsing with water and drying. This prior art involves an enzyme in addition to the dyeing material and a mordanting agent for colouring the hair.

US5447538 by Rosenbaum et al, teaches a milk or water-dilutable composition in dry form for application in diluted form to human hair to color said hair consisting essentially of a mixture of:(a) between 3 and 95 weight percent based on the total weight of said composition of particles of a vegetable substance wherein at least 95 percent by weight of said particles have a granulometry below 180 microns, said vegetable substance being of
20 the non-exhausted type and not naturally containing dye principles, said substance being selected from the group consisting of (1) the entire plant of wheat, (2) the leaves of a plant selected from the group consisting of cassia obovata, and (3) corn stalks,
(b) a direct dye in an amount of 0.05 to 30 percent by weight based on the total weight of said composition, said direct dye being selected from the group consisting of lawsone,
25 hematoxylin, purpurin, alizarin, indigo and curcumin, and
(c) a solid diluent present in an amount of 5 to 75 percent by weight based on the total weight of said composition, said solid diluent exhibiting in a 40% solution or dispersion in water a viscosity, at ambient temperature, not greater than 150 centipoises, said diluent being selected from the group consisting of powdered milk, glucose, levulose, lactose,
30 sorbitol, maltose, sucrose, starch, sodium carbonate, and sodium citrate.

This prior art mentions a vegetable substance that imparts to the composition good unctuousness after dilution, facilitates its removal and imparts both softness and a shiny appearance to the hair but does not refer to any mordanting agent in the composition.

US5725603 by Audousset et al., teaches about a process for the direct dyeing of human keratinous fibers which comprises directly dyeing the fibers by contacting the fibers with a composition containing at least one natural dye and with a gas containing water vapor, the temperature of the gas being at least 85-150 degree. C. and the contact time between the gas and the fibers to be dyed not exceeding two minutes. The hair is dyed in the absence of any mordanting agent in this prior art but using water vapour.

US5865853 by Schmitt et al., is directed to a composition for dyeing keratin fibers, said composition comprising from 35 to 75 percent by weight of at least one powdery vegetable dye material; from 25 to 65 percent by weight of at least one oil; and from 0.001 to 15 percent by weight of at least one synthetic direct-dyeing dye compound selected from the group consisting of nitro dye compounds, azo dye compounds, quinone dye compounds and triphenylmethane dye compounds. To adjust the pH of the dye-containing suspension according to this prior art, alkalizing agents, such as alkali metal hydroxides, sodium carbonates, sodium hydrogen carbonates, magnesium carbonates, ammonium carbonates, ammonium hydrogen carbonates or sodium silicates, or acidifying agents, such as citric acid, tartaric acid, ammonium chloride or ammonium sulfate are added. This prior art does not involve any mordanting agent but involves higher wt % of the dye material in the hair dyeing composition.

"Structure and properties of hematein derivatives" by Azuko Shirai and Masaru Matsuoka 1996. Dyes and pigments Vol: 32:3, relates to the investigation of oxidation of hematoxylin to hematein under various conditions, and the formation and absorption spectra of the hematein metal complexes were studied and conformational structures of hematoxylin and hematein were calculated by MOPAC PM3. The possibilities of their structural isomers, heat of formation, and their-dimensional structures were evaluated and the most stable conformers. This prior art does not hint at a hair dyeing composition but teaches on hematein metal complexes.

The above state of the art related to colouring/ dyeing of keratinous fibre reveal that while there are several processes for achieving such colouring, when use of mordanting agent is involved for such dyeing of keratinous fibers, to achieve the base colour of the dye there has been a necessity to involve additional additives in the nature of pH modulators/activators or oxidising agents. While such approach to generation of dye colour involving mordanting agent has been followed over the years, the nature of colouring obtained following such processes with additional additives as mentioned above was not

studied to the desired level and there has thus been a need to appreciate the function of pH regulators and/ or oxidising agents in such dyeing processes under ambient temperature conditions and to provide avenues for dyeing avoiding the use of pH regulators and/ or oxidising agents so that any unwanted harmful effects of such additional additives could be avoided. Moreover, the above state of the art further reveals the limitation of such known prior processes in generating variants of base colour involving only mordanting agents and dye at ambient temperatures.

OBJECTS OF THE INVENTION

It is thus the basic object of the present invention to explore the heretofore unknown possibilities of achieving ready and effective dyeing of keratinous fibres avoiding the use of external additives such as pH regulators, oxidizing agents to make dyeing of keratinous fibres more safe and convenient for variety of dye based applications under ambient temperature conditions.

Another object of the present invention is to provide for a dye/ plant dye/concentrated plant dye based hair colouring composition/ kit involving simply a mordanting agent and a dye adapted for generating a safe modified base colour and/ or safe base colour of the dye/ plant dye for effective and safe colouring/ dyeing of the keratinous fibers such as hair at ambient temperature avoiding any oxidizing and other external pH controlling agents.

Another object of the present invention is to provide for the said dye/ plant dye/ concentrated plant dye based hair colouring composition/ kit simply including selective metal salts/ mordants and dyes at their selective levels adapted to generate selectively modified base colour of the dye/ plant dye or safe base colour of the dye/ plant dye to thereby provide for extensive colour variance/ shades at ambient temperatures.

Yet another object of the present invention is to provide for the said composition/ kit comprising the said selective mordanting agent and the dye/ plant dye/ concentrated plant dye based hair colour in combination with other hair care benefiting agents, other compatible carriers of the said dyes and mordants to provide favouring different acceptable forms of the said composition/ kit including creams, lotions, powders for effective end use/ application in colouring/dyeing of the keratinic fiber under ambient temperature conditions.

It is yet another object of the present invention to provide for a dye/ plant dye/concentrated plant dye based hair colouring composition/ kit involving mordanting agent and said dye adapted to generate variety of colours just by changing the metal mordant wherein such wide variety of colour generation while colouring/ dyeing of the keratinous fibers would be achievable in a single step under ambient temperature conditions.

SUMMARY OF THE INVENTION

Thus according to the basic aspect of the present invention there is provided a hair colour composition/ kit for dyeing keratin fibers comprising selectively only atleast one or more dye, plant dye, concentrated plant dye and-atleast one or more inorganic metal salt based mordanting agent for generating dye/plant dye /concentrated plant dye-metal complex free of any pH controlling agents and oxidizing agents, with safe modified base colour and/ or base colour of the dye/ plant dye/ concentrated plant dye under ambient temperature conditions.

According to another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising:

- a) said atleast one or more dye, plant dye, concentrated plant dye;
- 20 b) said atleast one or more inorganic metal salt based mordanting agent and selectively anyone or more of
 - i) copper salts;and
 - ii) suitable hair grooming/ benefiting agent;
 - iii) an aqueous solution comprising anyone or more of lactic acid, IPA (iso propyl alcohol), Benzyl alcohol;-
- 25 iv) instructions for producing the safe colouration by generation of modified base colour and/ or base colour of the dye/plant dye/ concentrated plant dye including shine-metallic lustre to said keratinic fiber.

30 It is thus the surprising and selective finding of the present invention that by simply involving -at least one or more inorganic metal salt based mordanting agent in combination with a-at least one or more dye, plant dye, concentrated plant dye for generating dye/plant dye/ concentrated plant dye-metal complex free of any pH controlling agents and oxidizing agents, a safe modified base colour and/ or base colour of the dye/plant dye/ concentrated

plant dye could be achieved with variants of base colour shades with superior colour delivery at ambient temperature conditions

Advantageously, the delivery of variants of base colour shade and/ or base colour by the composition/ kit of the present invention could be thus readily achieved in a single step under ambient temperature conditions by selective inorganic metal salt without requiring the need of any pre-mordanting or post-mordanting steps for colour delivery.

According to another preferred aspect of the present invention there is provided the said hair colouring composition/ kit wherein said (a) dye/ plant dye/ concentrated plant dye is present in amounts of 0.1 to 15% and said (b) inorganic metal salt based mordanting agent is present in amounts of 0.01 to 10 percent of the active metal in the composition.

According to yet another preferred aspect of the present invention there is provided the said hair colouring composition/ kit wherein said (a) dye/ plant dye/ concentrated plant dye is either pure dye/ pure natural dye or an extract/ concentrate of the plant material or an unprocessed plant material that selectively includes flavonoids, terpenoids, polyphenolics or its derivatives as the basic molecular skeleton including selective additional monoazo, hydroxyl, carboxyl or amino groups and preferably comprises anthocyanins preferably Haematoxylin, Tanins, Benzoquinones, Naphthoquinones preferably Henna, Walnut, Anthroquinones preferably Alizarin, carotenoids, indigo, indigocaramine, rubiethyric acid, purpuroxanthin, rubiadin, morindanigrin, munjistin, morindadiol, carotene, crocentin, bixin, canthaxanthin, lycopene, capsanthin, apocarotenal, xanthophyll, curcumin, morin, malclurin, luteolin, apigenin, fukugetin, datiscetin, kaempferol, rhamnocitrin, rhamnethin, zanthorhamnin, isorhamnetin, rhammazin, quercetin, rutin, gossypetin, butin, rotterin, chlorophyll A & B, catechin, fisetin, lapachol, juglone, alkannin, alkannan, deoxysantalol, atromentin, awobamin, riboflavin, anthocyanin, lawsone, embilica extract, carajuirin, dracorbohdin, berberine, betanin orcein, xanthone, naphthalene, alfalfa extract, black tea extract, green tea extract, white tea extract and red sandalwood; and

wherein said (b) mordanting agent selectively includes inorganic metal salts including Ferrous sulphate, Ferric oxide, Ferric nitrate Zinc sulphate, Zinc nitrate, Zinc Chloride, Zinc Oxide, Copper Sulphate, Copper Nitrate, Copper Chloride, Copper Oxide, Magnesium Sulphate, Magnesium Nitrate, Magnesium Chloride, , Magnesium Oxide, Magnesium carbonate, Sodium Chloride, Sodium bicarbonate, Sodium Carbonate, Cobalt Nitrate, Tin Chloride, Barium Chloride, Lead Nitrate, Potassium tellurate.

According to another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising (a) haematoxylin and anyone or more mordanting agent (b) selected from ferrous chloride, ferrous sulphate.

5

According to yet another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising

(a) haematoxylin and anyone or more mordanting agent (b) selected from zinc sulphate, zinc chloride, zinc nitrate, zinc oxide, potassium tellurate; and (a) Alizarin and anyone mordanting agent (b) selected from magnesium oxide, magnesium carbonate, sodium carbonate, potassium tellurate.

10

According to another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising

(a) haematoxylin and anyone or more mordanting agent (b) selected from copper sulphate, copper nitrate, copper chloride.

15

According to yet another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising

(a) Alizarin and anyone or more mordanting agent (b) selected from zinc sulphate, zinc chloride, zinc nitrate, zinc oxide.

20

According to another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising

(a) Quercetin and anyone or more mordanting agent (b) selected from zinc chloride, zinc nitrate, magnesium sulphate, magnesium chloride, magnesium nitrate, magnesium oxide.

25

According to yet another preferred aspect of the present invention there is provided the said hair colour composition/ kit comprising

(a) alizarin and anyone or more mordanting agent (b) selected from magnesium carbonate, magnesium chloride, sodium carbonate, sodium chloride.

30

According to another aspect of the present invention there is provided the said hair colour composition/ kit involving a hair colour formulation in at least two parts comprising the

basic essential ingredients of (a) atleast one or more dye, plant dye, concentrated plant dye and (b) atleast one or more inorganic metal salt based mordanting agent.

According to yet another aspect of the present invention there is provided the said hair colour composition/ kit additionally comprising hair colour/ dye/ plant dye/ mordanting agent acceptable carriers involving emulsifiers⁴⁶ selected from anyone or more of anionic, cationic, non-ionic and amphoteric surfactants; and also comprises suitable hair benefiting agents and color enhancing diffusion aids present in selective levels.

According to yet another aspect of the present invention there is provided the said hair colour composition/ kit suitably made available in various physical forms including powders, creams, lotions, sprays, mousse, gel.

According to another aspect of the present invention there is provided a method of colouring the hair involving said hair colouring composition/kit comprising the steps of:

- a. providing an aqueous and/or alcoholic solution of the said inorganic metal salt based mordanting agent with or without hair benefiting/conditioning agents on hair;
- b. applying said dye molecule on the hair in solid and/ or in solution form favouring complex formation on the surface or within the hair for effective colouration of the hair through generation of safe modified base colour and/ or safe base colour of the dye/plant dye/concentrated plant dye including imparting shine-metallic lustre to said keratinic fiber under ambient temperature conditions.

According to yet another preferred aspect of the present invention there is provided a method of colouring the hair involving hair colouring composition/kit comprising the steps of:

- a. mixing of the inorganic metal salt based mordanting agent with or without hair benefiting/conditioning agents and dye/ plant dye/concentrated plant dye with or without hair benefiting/conditioning agents to achieve uniform consistency;
- b. applying the mixture to hair/keratin fibre favouring effective colouration of keratinous fibres in variety of shades under ambient temperature conditions.

Yet another preferred aspect of the present invention there is provided a composition/ kit for dyeing keratin fibres wherein the metal ion solution pH is the natural intrinsic pH of the mordant solution under which it effectively complexes with the dye to yield different insoluble metal ion-dye complex delivering different colours.

The invention is illustrated hereunder in greater detail in relation to the non-limiting exemplary illustration hereunder:

5 DETAILED DESCRIPTION OF THE INVENTION

As discussed hereinbefore, the present invention provides for a hair colour composition/ kit for dyeing keratin fibers comprising selectively atleast one or more dye, plant dye, concentrated plant dye in combination with atleast one or more inorganic metal salt based mordanting agent for generating dye/ plant dye/concentrated plant dye-metal complex free of pH controlling agents and oxidizing agents, with safe modified base colour and/ or base colour of the dye/ concentrated plant dye thereby displaying superior colour delivery and colour variance under ambient temperature conditions.

15 Preferably, the said dye/plant dye/ concentrated plant dye based hair colour composition/ kit provides for complexed dye/plant dye/ concentrated plant dyes with inorganic metal salt based mordants wherein the dyes/ plant dyes favour generation of the modified and/ or base colour of the said dye/plant dye when present in their selective levels.

20 The said dye/ plant dye/ concentrated plant dye molecules include anyone or more flavonoids, terpenoids polyphenolics or its derivatives, as the basic molecular skeleton of the said dye/ plant dye and preferably comprises anthocyanins (such as Haematoxylin), Tanins, Benzoquinones, Naphthoquinones (such as Henna, Walnut), Anthroquinones (such as Alizarin), & carotenoids including selective additional monoazo, hydroxyl, carboxyl or amino groups, which are capable of forming strong co-ordination complexes with transition metal ions such as chromium, cobalt, nickel, copper and the like.

EXAMPLE I: REACTION PROTOCOL

30 The experiment protocol is based on conventional methodology for the generation of modified base colour and/ or base colour shades of the dyes involving dye/ plant dye/ concentrated plant dye combination with at least one or more inorganic metal salt based mordanting agent and the results are summarized in the following **Table 1**.

Table 1

35

S. No.	Treatment Details	Shade
<u>Black shades</u>		
1	Haematoxylin + 3% Ferrous chloride	Brownish Black shade
2	Haematoxylin + 3% Ferrous sulphate	Natural black
<u>Violet shades</u>		
3	Haematoxylin + 3% zinc sulphate	Violet shade
4	Haematoxylin + 3% zinc chloride	Violet shade
5	Haematoxylin + 3% zinc nitrate	Violet shade
6	Haematoxylin + 3% zinc oxide	Dark violet shade
7	Alizarin + 3 % Magnesium oxide	Dark violet shade
8	Alizarin + 3 % Magnesium carbonate	Dark violet shade
9	Alizarin + 3 % Sodium carbonate	Dark violet shade
10	Haematoxylin + 3 % potassium tellurate	Violet shade
11	Alizarin + 3 % potassium tellurate	Dark violet shade
<u>Brown shades</u>		
12	Haematoxylin + 3% copper sulphate	Brown
13	Haematoxylin + 3% cupric nitrate	Metallic brown
14	Haematoxylin + 3% cupric chloride	Very shine metallic brown
15	Haematoxylin + 3% cupric acetate	brown
<u>Burgundy shades</u>		
16	Alizarin + 3 % zinc sulphate	Dark burgundy
17	Alizarin + 3 % zinc chloride	burgundy
18	Alizarin + 3 % zinc nitrate	burgundy
19	Alizarin + 3 % zinc oxide	burgundy
<u>Yellow shades</u>		
20	Quercetin + 3 % zinc chloride	Dark yellow
21	Quercetin + 3 % zinc nitrate	Dark yellow
22	Quercetin + 3 % Magnesium sulphate	Yellow
23	Quercetin + 3 % Magnesium chloride	Yellow
24	Quercetin + 3 % Magnesium nitrate	Yellow
25	Quercetin + 3 % Magnesium oxide	Yellow

Hence it is concluded from the above table, that the dye haematoxylin which originally has an ash shade, also generates different modified colour shades simply upon selective combination with specific inorganic metal salt based mordanting agents in the absence of any pH controlling agent or oxidizing agent.

- 5 It is further observed from the above table that Iron salts reacted with Haematoxylin produce the black/ brown shade, Zinc salts react with alizarin to generate burgundy shade, copper salts react with haematoxylin to yield the brown shade, salts of magnesium reacted with Quercetin to yield yellow shades, Zinc salts reacted with haematoxylin to generate violet shades, Potassium tellurate reacted with haematoxylin to yield violet shade and also
 10 reacted with alizarin to generate dark violet shade, magnesium salts and sodium salts also reacted with alizarin to result in dark violet/ violet shades thus displaying a broad colour variance in the absence of any pH controlling agent and oxidizing agent.

15 **EXAMPLE II:** The effect of pH controlling agent on the colour of inorganic metal mordant-dye complex:

Table 2

S. No	Details	pH of the metal salt solution	Treatments	Shade
DATA SET I				
1	1.0 % Haematoxylin + 3.0 % FeCl ₂	1.67	-	Brownish black shade
2	1.0 % Haematoxylin + 3.0 % FeSO ₄	5.18	-	Natural black shade
3	1.0 % Haematoxylin + 3.0 % FeSO ₄	1.49	Aqua acid	No dye uptake
4	1.0 % Haematoxylin + 3.0 % FeSO ₄	1.69	Lactic acid	No dye uptake
5	1.0 % Haematoxylin + 3.0 % Ferrous gluconate	4.76	-	Dull blue shade
DATA SET II				
1	1.0 % alizarin + 3.0 % Magnesium carbonate	11.87	-	Dark violet shade

2	1.0 % alizarin + 3.0 % Sodium carbonate	11.89	-	Dark violet shade
3	1.0 % alizarin + 3.0 % Magnesium Chloride	6.73		violet shade
4	1.0 % alizarin + 3.0 % Magnesium Chloride	11.89	Ammonia (NH ₃)	No dye uptake
5	1.0 % alizarin + 3.0 % Magnesium Chloride	11.21	Mono ethanol amine (MEA)	No dye uptake
6	1.0 % alizarin + 3.0 % Magnesium acetate	7.11	-	Dull violet shade

It is noted from Table 2 that the combination of ferrous chloride and haematoxylin yielded a brownish black shade under intrinsic ferrous chloride solution pH of 1.67. Therefore the intrinsic pH of ferrous sulphate, involving a different ferrous salt was also adjusted with external acidic agent like lactic acid and aqua acid to desired similar level of 1.6 to explore the reasons for wide colour variance that may be contributed by specific inorganic metal salt based mordanting agent. The dye uptake and shade direction are summarized as follows:

It was indeed found from table 2 above that the dye uptake was worst in the case of pH adjusted swatches. The swatch analysis revealed that the colour delivery was influenced solely by specific inorganic metal salt mordants and not influenced at all by pH of the mixture. Therefore the experiment proved that the shade direction was not influenced by pH and is influenced only by the specific inorganic metal salt mordant.

Similarly experiment was carried out in alkaline range. The magnesium carbonate and sodium carbonate yielded the dark violet shade with alizarin. The pH range of sodium carbonate and magnesium carbonate is 11.8. Therefore the pH of magnesium chloride was adjusted with ammonia and mono ethanolamine (MEA) and no dye uptake was observed. The above study thus confirmed the role of specific inorganic metal salt mordants to be effective in displaying such wide colour variance and therefore the colour variance is essentially and simply based on selective at least one or more of inorganic metal salt based mordanting agent in combination with at least one or more dye, plant dye, concentrated plant dye for generation of specific colour shades including modified base colour of the said dye thereby providing for wide colour variance.

More specifically the above table also clearly illustrates that the inorganic metal salt based mordanting agents provides for superior colour delivery over the organic acid salts.

Thus it is evident from the results obtained under tables 1 and 2 that the technical advance of the hair dyeing composition/ kit made possible by way of the present invention is based on the surprising finding of a selective combination of selectively only one-atleast one or more dye, plant dye, concentrated plant dye and one-atleast one or more inorganic metal salt based mordanting agent for generating dye/plant dye/ concentrated plant dye-metal complex free of any pH controlling agents and oxidizing agents, with safe modified base colour and/ or base colour of the dye/ concentrated plant dye, for effective and safe colouration of keratinous fibres including shine-metallic luster under ambient temperature conditions.

Characteristic features of the hair dyeing composition/kit of the present invention and the advantages obtained as set forth hereunder:

- a) A hair dyeing composition/kit simply comprising atleast one or more dye, plant dye, concentrated plant dye in combination with one-atleast one or more inorganic metal salt based mordanting agent for generating safe modified base colour and/ or safe base colour in the absence of oxidizing and other external pH controlling agents, for effective colouration of the keratinous fibers under ambient temperature conditions.
- b) A hair dyeing composition/kit comprising selectively only at least one or more dye, plant dye, concentrated plant dye in combination with one-atleast one or more inorganic metal salt based mordanting agent including selective inorganic metal salts/ mordants at their selective levels adapted to generate selective modified base colour and/or base colour of the dye/ plant dye to thereby provide for extensive colour variance/ shades without requiring any pre-mordanting and post-mordanting steps for said generation of extensive colour shades in a single step under ambient temperature conditions.
- c) A natural hair dyeing composition/kit provides a much simple and cost-effective route for the purpose of dyeing of keratin fibers.

WE CLAIM:

1. A hair colour composition/ kit for dyeing keratin fibers comprising selectively atleast one or more dye, plant dye, concentrated plant dye and atleast one or more inorganic metal salt based mordanting agent for generating dye/concentrated plant dye-metal complex free of any pH controlling agents and oxidizing agents, with safe modified base colour and/ or base colour of the dye/ concentrated plant dye under ambient temperature conditions.
2. A hair colour composition/ kit as claimed in claim 1 comprising:
- a) said at least one or more dye, plant dye, concentrated plant dye;
 - 10 b) said at least one or more inorganic metal salt based mordanting agent and selectively anyone or more of
 - i) copper salts;
 - ii) suitable hair grooming/ benefiting agent;
 - iii) an aqueous solution comprising anyone or more of lactic acid, IPA (iso-propyl alcohol), Benzyl alcohol;
 - 15 iv) instructions for producing the safe colouration by generation of modified base colour and/ or base colour of the dye/plant dye/concentrated plant dye including shine-metallic lustre to said keratinic fiber.
3. A hair colour composition/ kit as claimed in anyone of claims 1 to 2 wherein said (a) dye/ plant dye/ concentrated plant dye is present in amounts of 0.1 to 15% and said (b) inorganic metal salt based mordanting agent is present in amounts of 0.01 to 10 percent of the active metal in the composition.
4. A hair colour composition/ kit as claimed in anyone of the preceding claims
- 25 wherein said (a) dye/ plant dye/ concentrated plant dye is either pure dye/ natural dye or an extract/ concentrate of the plant material or an unprocessed plant material that selectively includes flavonoids, terpenoids, polyphenolics or its derivatives as the basic molecular skeleton including selective additional monoazo, hydroxyl, carboxyl or amino groups and preferably comprises anthocyanins preferably Haematoxylin, Tanins, Benzoquinones, Naphthoquinones preferably Henna, Walnut, Anthroquinones preferably Alizarin, carotenoids, indigo, indigocaramine, rubiethyric acid, purpuroxanthin, rubiadin, morindanigrin, munjistin, morindadiol, carotene, crocentin, bixin, canthaxanthin, lycopene, capsanthin, apocarotenal, xanthophyll, curcumin, morin, malclurin, luteolin, apigenin, fukugetin, datiscetin, kaempferol, rhamnocitrin, rhamnethin, zanthorhamninn, isorhamnnetin,
- 30 rhammazin, quercetin, rutin, gossypetin, butin, rotterin, chlorophyll A & B, catechin, fisetin,
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lapachol, juglone, alkannin, alkannan, deoxysantalol, atromentin, awobamin, riboflavin, anthocyanin, lawsone, embilica extract, carajuirin, dracorbohadin, berberine, betanin orcein, xanthone, naphthalene, alfalfa extract, black tea extract, green tea extract, white tea extract and red sandalwood; and

5 wherein said (b) mordanting agent selectively includes inorganic metal salts including Ferrous sulphate, Ferric oxide, Ferric nitrate, Zinc sulphate, , Zinc nitrate, Zinc Chloride, Zinc Oxide, Copper Sulphate, Copper Nitrate, Copper Chloride, Copper Oxide, Magnesium Sulphate, Magnesium Nitrate, Magnesium Chloride, Magnesium Oxide, Magnesium carbonate, Sodium Chloride, Sodium bicarbonate, Sodium Carbonate, Cobalt Nitrate, Tin
10 Chloride, Barium Chloride, Lead Nitrate, Potassium tellurate.

5. A hair colour composition/ kit as claimed in anyone of the claims 1-4 comprising (a) haematoxylin and anyone or more of mordanting agent (b) selected from ferrous chloride, ferrous sulphate.

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6. A hair colour composition/ kit as claimed in anyone of the claims 1-4 comprising (a) haematoxylin and anyone or more mordanting agent (b) selected from, zinc sulphate, zinc chloride, zinc nitrate, zinc oxide, potassium tellurate; and (a) Alizarin and anyone mordanting agent (b) selected from magnesium oxide, magnesium carbonate, sodium
20 carbonate, potassium tellurate.

7. A hair colour composition/ kit as claimed in anyone of claims 1-4 comprising (a) haematoxylin and anyone or more mordanting agent (b) selected from, copper sulphate, copper nitrate, copper chloride.

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8. A hair colour composition/ kit as claimed in anyone of claims 1-4 comprising (a) Alizarin and anyone or more mordanting agent (b) selected from, zinc sulphate, zinc chloride, zinc nitrate, zinc oxide.

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9. A hair colour composition/ kit as claimed in anyone of claims 1-4 comprising (a) Quercetin and anyone or more mordanting agent (b) selected from zinc chloride, zinc nitrate, magnesium sulphate, magnesium chloride, magnesium nitrate, magnesium oxide.

10. A hair colour composition/ kit as claimed in anyone of claims 1-4 comprising

(a) alizarin and anyone or more mordanting agent (b) selected from magnesium carbonate, magnesium chloride, sodium carbonate, sodium chloride.

11. A hair colour composition/ kit as claimed in anyone of claims 1-4 involving a hair colour formulation in at least two parts comprising the basic essential ingredients of (a) at least one or more dye, plant dye, concentrated plant dye and (b) at least one or more inorganic metal salt based mordanting agent.

12. A hair colour composition/ kit as claimed in anyone of claims 1-4 additionally comprises hair colour/ dye/ plant dye/ mordanting agent acceptable carriers involving emulsifiers selected from anyone or more of anionic, cationic, non-ionic and amphoteric surfactants; and also comprises suitable hair benefiting agents and color enhancing diffusion aids present in selective levels.

13. A hair colour composition/ kit as claimed in anyone of claims 1-4 is suitably made available in various physical forms including powders, creams, lotions, sprays, mousse, gel.

14. A method of colouring the hair involving hair colouring composition/kit as claimed in anyone of claims 1-4 comprising the steps of:

a. providing an aqueous and/or alcoholic solution of the inorganic metal salt based mordanting agent with or without hair benefiting/conditioning agents on hair;

b. applying the dye molecule on the hair in solid and/ or in solution form favouring complex formation on the surface or within the hair for effective colouration of the hair through generation of safe modified base colour and/ or safe base colour of the dye/plant dye/ concentrated plant dye including imparting shine-metallic lustre to said keratinic fiber under ambient temperature conditions.

15. A method of colouring the hair involving hair colouring composition/kit as claimed in anyone of claims 1-4 comprising the steps of:

a. mixing of the inorganic metal salt based mordanting agent with or without hair benefiting/conditioning agents and dye/ plant dye/concentrated plant dye with or without hair benefiting/conditioning agents to achieve uniform consistency;

b. applying the mixture to hair/keratin fibre favouring effective colouration of keratinous fibres in variety of shades under ambient temperature conditions.

16. A hair colour formulation for dyeing keratin fibers and a hair colour composition/ kit obtained thereof substantially as herein described and illustrated with reference to the accompanying examples.

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INTERNATIONAL SEARCH REPORT

International application No. PCT/IN2012/000173
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<p>A. CLASSIFICATION OF SUBJECT MATTER IPC(8) - A61Q 5/10 (2012.01) USPC - 8/405 According to International Patent Classification (IPC) or to both national classification and IPC</p>
<p>B. FIELDS SEARCHED</p> <p>Minimum documentation searched (classification system followed by classification symbols) IPC(8) - A61Q 5/06, 08, 12 (2012.01) USPC - 8/407; 8/425; 8/428; 8/429; 8/435; 8/646</p> <p>Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched</p> <p>Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) PatBase, Orbit, Google Patents, Google</p>

C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	IN 2513/CHE/2008 (RADHAKRISHNAN et al) 23 March 2010 (23.03.2010) entire document	1-3
Y	CN 101812246 A (LIU et al) 25 August 2010 (25.08.2010) entire document	1-3
A	US 2010/0154140 A1 (SIMONET et al) 24 June 2010 (24.06.2010) entire document	1-3
A	US 2010/0306929 A1 (DE BONI et al) 09 December 2010 (09.12.2010) entire document	1-3

Further documents are listed in the continuation of Box C.

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|---|---|
| <p>* Special categories of cited documents:</p> <p>“A” document defining the general state of the art which is not considered to be of particular relevance</p> <p>“E” earlier application or patent but published on or after the international filing date</p> <p>“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)</p> <p>“O” document referring to an oral disclosure, use, exhibition or other means</p> <p>“P” document published prior to the international filing date but later than the priority date claimed</p> | <p>“T” later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>“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</p> <p>“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</p> <p>“&” document member of the same patent family</p> |
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Date of the actual completion of the international search 10 August 2012	Date of mailing of the international search report 31 AUG 2012
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Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-3201	Authorized officer: Blaine R. Copenheaver PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/IN2012/000173

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.: 16
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
Claim 16 has been held as an omnibus claim, as it refer to an invention "substantially as described and illustrated with reference to the accompanying examples."

3. Claims Nos.: 4-15
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- No protest accompanied the payment of additional search fees.