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[54] **PLANOGRAPHIC REVERSED PRINTING**
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451, 452, 452—473

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ABSTRACT: A process for producing planographic prints wherein the printing plate is prepared from a heat-sensitive recording material formed of a support, an interlayer of a water-permeable hydrophilic colloid and an external water-permeable layer of at least one hydrophobic substance which melts on heating and in such molten condition is absorbed into the hydrophilic colloid interlayer by exposing such recording material to a pattern of heat to bring about differential absorption of the hydrophobic substance in the heated regions of the layer thereof to thereby expose portions of the hydrophilic layer in a pattern corresponding to the heat pattern. The planographic plate thus produced is inked with a generally hydrophilic printing ink selectively adhering to the hydrophilic portions, and the ink is transferred to a copy material to produce the desired print. Preferably, the hydrophobic substance is miscible or compatible in its molten state with the hydrophilic colloid of the interlayer. A particularly useful printing ink consists of a colored hydrophilic continuous phase in which is dispersed a lipophilic phase of coloration distinct from that of the hydrophilic phase.

PLANOGRAPHIC REVERSED PRINTING

The present invention relates to planographic printing and particularly to the use of planographic printing masters the printing image of which is hydrophilic.

Planographic printing is based on the physical property of repellence of greasy materials to water. In ordinary planographic printing the printing master surface contains the pattern of the image to be printed in terms of a differentiation in water repellency. The printing plate is usually prepared by imagewise affixing of a water repellent substance or composition, usually greasy, resinous or waxy in nature to a hydrophilic surface. According to a more recent technique a hydrophilic surface is imagewise converted into a hydrophobic one, by heat absorbed in a thermographic recording material as is described, e.g., in the Belgian Pat. specification 656,713 and the published Dutch Pat. application 6,606,719.

In ordinary planographic printing use is made of inks that are composed of a lipophilic phase wherein pigments and other suitable ink ingredients are worked up, which do not absorb water or only to a small extent. During printing according to the classical planographic process the printing plate is kept supplied with an aqueous composition so as to keep the non-printing areas sufficiently hydrophilic. This method of printing thus requires in addition to an inking system a so-called "damping system" by means of which the hydrophilic areas are covered with water or a colorless aqueous composition and thus kept grease repellent.

According to a more recent developed planographic printing process, called "reversed planographic printing process" use is made of a printing master, wherein the hydrophilic parts constitute the printing parts. The printing ink used in this process contains an aqueous ink composition as colored medium and the damping system is used for applying a colorless oleophilic liquid composition. The reversed planographic printing can be carried out as "direct planographic printing" in other words planographic printing wherein no image reversal takes place (offset) by means of an intermediate cylinder with rubber blanket.

It is one of the objects of the present invention to provide a printing process wherein use is made of a printing master the printing parts of which are made hydrophilic with the aid of heat and/or pressure.

It is a particular object of the present invention to use in a direct planographic printing process a printing master having a laterally reversed hydrophilic printing image, and wherein the said hydrophilic image is obtained by imagewise applied heat and/or pressure to a recording element (printing master blank) having an hydrophobic surface layer which is applied to a hydrophilic or hydrophilizable interlayer or support.

A process has now been found for the production of prints by means of a printing master containing the step of forming a pattern of hydrophilic portions in a hydrophobic thermoplastic surface layer of a recording material by imagewise or recordwise heating or imagewise or recordwise exerting pressure to the hydrophobic surface layer of said material, wherein said surface layer has been applied to a hydrophilic or hydrophilizable interlayer or support.

More particularly it has been found that a reversed planographic printing master can be produced by a process containing the steps of heating a recording material, which comprises a thermoplastic hydrophobic surface layer applied to a hydrophilic or hydrophilizable interlayer or support, in accordance with the image to be reproduced, or imagewise or recordwise applying pressure to said recording material and imagewise or recordwise removing the heated portions of said surface layer, and/or allowing said heated portions to penetrate in the hydrophilic or hydrophilizable interlayer or support.

The penetration of the heated portions of the hydrophobic thermoplastic surface layer in the hydrophilic interlayer or support can be realized by using a porous hydrophilic interlayer or support to which the hydrophobic surface layer is applied as a thin film or can be realized by using hydrophobic melttable substance(s) in the surface layer which on heating

become(s) compatible with (dissolve(s) in) the hydrophilic substances of the interlayer or support.

According to a first embodiment for producing a planographic printing master suited for reversed planographic printing a hydrophobic surface layer containing hydrophobic thermoplastic or sublimable substances, which have been applied to a hydrophilic or hydrophilizable interlayer or support, is imagewise removed therefrom and transferred to the image markings of an original by infrared irradiating the original while in contact with the said surface layer and subsequently separating the latter from the original. The said selective transfer is the result of the imagewise melting, softening or sublimation of the thermoplastic hydrophobic substances of the surface layer which during the irradiation is in contact with the heated image markings of the original. For carrying out that embodiment, preferably a recording material with a hydrophobic surface layer having a thickness of 0.1μ to 20μ is used.

In order to prevent the transfer of the hydrophobic substances to the original, thus in order to avoid its smudging, the surface recording layer may be covered with an intermediate sheet (auxiliary sheet), which during the exposure is arranged between the original and the heat-sensitive surface layer. Said intermediate sheet, which preferably has a porous structure, e.g. is a thin fat-absorbing paper sheet, and is heat conductive in the embodiment wherein the heat is produced in the image markings of the original.

In an embodiment described further on wherein heat is imagewise generated in radiation-absorbing substances contained in the thermoplastic hydrophobic surface recording layer, the said intermediate sheet preferably is nonheat conductive (prevents conduction of heat from the original) but is transparent for the heat-generating radiation used in the exposure.

According to a second embodiment for preparing a planographic printing master suited for reversed planographic printing, portions of a hydrophobic thermoplastic surface layer are melted in accordance with heated image markings of an original and in molten state dissolved in a suitable underlying hydrophilic layer or support. For that purpose the hydrophobic substances of the surface layer are chosen in such a way that the molten hydrophobic substances become compatible with (in other words dissolve in) the hydrophilic composition of said interlayer or support.

According to a third embodiment for preparing a planographic printing master suited for reversed planographic printing, portions of a thermoplastic hydrophobic surface layer are melted in accordance with heated image markings of an original and in molten state absorbed in the porous structure of an underlying hydrophilic support or interlayer.

According to a fourth embodiment a planographic printing master suited for reversed planographic printing is produced by imagewise or recordwise application of pressure through a support of a recording material containing a hydrophobic pressure-sensitive surface layer coated on a hydrophilic or hydrophilizable surface of the support or an interlayer. By pressure sensitivity is meant that the hydrophobic layer is frangible and e.g. under the impact of the relief of the letter of a typewriter key is broken so that a corresponding part is transferred to a receiving material as in classical duplicating with a typewriter carbon paper. In the present embodiment a support for the pressure-sensitive material is chosen that is as thin as possible but is still sufficiently strong to withstand the mechanical forces during typewriting and printing.

In the embodiment wherein a printing master blank is used wherein the hydrophobic surface layer has been applied to a hydrophilizable interlayer or support i.e. an interlayer or support which is initially hydrophobic, and wherefrom by heat and/or pressure the hydrophobic surface layer is imagewise removed, the hydrophilization of that interlayer or support takes place in the areas that are no longer covered by the surface layer.

The said hydrophilization can be carried out after bracing the printing master on the printing cylinder and before starting printing.

According to a preferred technique said hydrophilization is carried out just before printing with the aqueous printing ink itself, which ink for that purpose contains hydrophilizing substance(s) for the hydrophilizable interlayer or support.

Specific examples of hydrophobic layers that can be hydrophilized according to techniques known by those skilled in the art, are e.g. so-called zinc oxide layers as used in electrophotography. Such layers containing photoconductive zinc oxide grains dispersed in an insulating hydrophobic binder can be made hydrophilic, e.g., by means of potassium hexacyanoferrate (III) and/or acid phosphates. Another example of an hydrophilizable hydrophobic sheet material is an anode-oxidized aluminum sheet, which by a treatment with a strong base is made hydrophilic and kept hydrophilic with phosphoric acid and hydrophilic colloids such as arabic gum (see for a suitable hydrophilization composition for aluminum sheets in the U.K. Pat. specifications 913,591 and 946,538). A further example of a hydrophilizable sheet material is a material containing a bimetal layer or sheet of which the metal coated with the hydrophobic surface layer is hydrophobic, e.g. copper, and the underlying metal is hydrophilic, e.g. chromium. After imagewise removal of the hydrophobic thermoplastic surface layer, the bare copper parts are etched away, e.g. with iron (III) chloride so that the uncovered chromium layer portions can act as hydrophilic ink-receptive printing parts.

Another example of a hydrophilizable sheet or layer is a cellulose triacetate sheet or layer, which can be hydrophilized by hydrolysis at its surface by an aqueous solution of a strong base.

Having stated in general the different embodiments and possible concepts of printing master blanks suited for use according to the present invention, a more detailed survey of the ingredients suitable for use in these printing master blanks will now be given.

In the preparation of the hydrophobic surface layer use is preferably made of a film-forming thermoplastic hydrophobic substance or mixture of substances, which is applied in such a way that a water-impermeable layer is obtained. Preferably a thermoplastic substance is used which has a melting point, melting range or sublimation range comprised between 40° and 120°C.

As sublimable hydrophobic compounds can be mentioned: biphenyl, camphor, and naphthalene.

As film-forming thermoplastic hydrophobic substances can be mentioned water-repellent substances of a waxlike consistency, which include natural, modified as well as synthetic waxes, or compounds that make part of the mixture of substances composing these waxes.

So, reference is made to hydrocarbons, fatty acids and the metal salts thereof, fatty alcohols, and higher aliphatic ketones, ethers and esters, and to sterols and hydrophobic derivatives thereof irrespective of their source or method of preparation.

The hydrophobic surface layer is preferably composed of or mainly contains (preferably more than 80 percent by weight) such substances.

In the embodiment wherein the hydrophobic substance has to be removed by adherence to the original or intermediate sheet or dissolved in the underlying hydrophilic element, preferably hydrophobic meltable substances having a low melt viscosity are used, e.g. waxes that are composed of long chain alkanes (C₁₈—C₃₀) and/or aliphatic compounds known as fatty acids, fatty alcohols, fatty esters and ethers.

Thus use can be made of known waxes such as natural waxes, e.g. insect waxes, animal waxes, and vegetable waxes; mineral waxes, e.g. petroleum waxes and analogous synthetic representatives.

The following is a nonlimitive list of waxes containing representatives suitable for use according to the present invention.

I. Natural waxes

a. insect waxes: e.g. beeswax, chinese insect wax, shellac wax;

b. animal waxes: e.g. spermacetic wax;

c. vegetable waxes: e.g. carnauba wax, ouricury wax, candellila wax, sugar cane wax.

II. Mineral waxes: e.g. ozocerite wax, Utah wax, peat wax, Montan wax and paraffin wax separated from petroleum.

III. Synthetic waxes: e.g. chemically modified natural waxes and mineral waxes e.g. the so-called "IG-waxes," chlorinated paraffin and synthetic paraffins prepared from ethylene.

Other synthetic waxes are, e.g., those sold under the trade-names ACRAWAX, ARMID and VOLTALEF. ACRAWAX is a registered trademark of Glyco Chemical, Inc. New York, N.Y., USA for complex nitrogen derivatives of the higher fatty acids. ARMID is a registered trademark of Armour Industrial Chemical Company, Chicago, Ill., U.S.A., for fatty acid amides. VOLTALEF is a registered trademark of Pechiney, St. Gobain, Paris, France, for chlorine- and fluorine-substituted hydrocarbon.

Detailed information about different types of waxes can be found in the above-mentioned book of Bennet H., and in "The Chemistry and Technology of Waxes," (by Albin H. Warth), 2nd ed. Reinhold Publishing Corp., New York.

It may be advantageous for the purpose of raising the melting point or lowering the melt viscosity, to employ e.g. a mixture of two or more waxlike materials in the recording layer.

For instance a certain amount of beeswax or carnauba wax can be mixed with paraffin wax to raise the melting point of the paraffin wax.

Another example is the addition of ozocerite wax, which is soft and plastic, to a paraffin wax of approximately the same melting point, which is hard and lacks plasticity. Softening of the wax can also be effected successfully by adding thereto oils such as paraffin oil or weak greasy substances such as petrolatum.

The hydrophobic surface recording layer having a non-porous structure i.e. a nonwater-permeable structure need not necessarily consist of or contain waxes, which means that hydrophobic thermoplastic polymers or resins can be used instead of waxes e.g. silicone resins, polyethylene, polyvinyl stearate, polyhexamethylene adipate, a hydrogenated glyceryl ester of stearic acid, or a polyester of sebacic acid and 1,6-hexane-diol.

Hydrophilic interlayers (when used) for absorbing molten hydrophobic substances preferably mainly contain hydrophilic colloids of the following classes:

A. water-soluble natural polymeric substances e.g. agar-agar, alginates, amylose, amylopectine, dextrine, gum arabic, casein and gelatin;

B. modified hydrophilic natural polymeric substances derived from starch, cellulose and alginic acid.

As water-soluble starch derivatives are mentioned:

1. nonionic starches:

acetates e.g. corn starch acetate containing 1 to 4 percent of acetyl groups;
starch hydroxyethyl ethers;
hydroxyalkyl-starch derivatives (e.g. hydroxyethyl- and hydroxypropyl-starch derivatives).

(the ionic starches:

(the introduction of carboxyl, sulfonate, sulfate, or phosphate groups into starch permits the preparation of water-soluble alkali metal or ammonium salts of anionic starches, whereas the introduction of amino groups permits the preparation of cationic starches e.g. amino-alkyl corn starch).

As water-soluble cellulose derivatives are mentioned:

1. nonionic cellulose gums:

(this category includes hydroxyalkyl-cellulose derivatives and polyoxyalkylene-cellulose derivatives).

2. ionic cellulose gums (the introduction of carboxyl, sulfonate sulfate or phosphate groups into cellulose permits the preparation of water-soluble alkali metal or ammonium salts of anionic cellulose derivatives).

Suitable ionic cellulose gums are e.g. carboxymethyl-cellulose methylcellulose-m-sulfobenzoate, ethylcellulose sulposuccinate and acetylcellulose sulfo succinate

C Synthetic water-soluble resins. Among these resins can be mentioned:

1. water-soluble homo- and co-vinyl polymers containing, e.g., vinyl alcohol, N-vinylpyrrolidinone, vinyl methyl ether, acrylic acid, methacrylic acid, maleic acid, and acryl-amide units.
2. water-soluble homo- and copolymers containing alkylene oxide units such as ethylene oxide units;
3. water-soluble polycondensates e.g. water-soluble alkyd resins and water-soluble polyester compounds.

The hydrophilic interlayers need not necessarily contain hydrophilic water-soluble polymers, or colloids but may contain hydrophilic water-permeable water-insoluble polymers e.g. may contain cellulose derivatives that contain a certain amount of hydrophilic substituents providing water-permeability such as partly etherified or esterified cellulose e.g., methylcellulose with low methoxy degree, ethylcellulose, benzylcellulose, hydroxyethylcellulose acetate, cellulose acetate sorbate, and cellulose acetate butyrate, partly hydrolyzed polyvinyl acetate, copolymers of styrene and allyl alcohol, copolymers containing unacetalized hydroxyl groups, and vinyl copolymers containing small amounts of carboxylic acid or carboxylic anhydride, e.g., maleic anhydride groups.

In order to determine, whether a hydrophobic substance can be dissolved in a given hydrophilic polymer or colloid, a method can be applied, which consists in determining the melting point of the hydrophobic fusible material alone and of the mixture of said fusible material with the hydrophilic polymer or colloid chosen. Prior to the determination of the melting point the mixture must have been heated beyond the melting point of the hydrophobic substance and then been cooled. In this test a substantial increase in the melting point of the said mixture beyond that of the fusible hydrophobic substance alone indicates that the substances are compatible, whereas a substantially unchanged melting point indicates a lack of compatibility. In this type of test the hydrophilic polymer alone has a melting or softening point higher than that of the hydrophobic fusible substance.

Another test which is of value to select compatible partners consists in heating a dispersion made at room temperature of the hydrophobic partner in the hydrophilic one. When the dispersion by heating is transformed into a transparent homogenous molecular mixture, and after being cooled remains more transparent than it was before heating, one can conclude that the partners are capable of forming a compatible mixture with each other.

Printing master blanks that are heat sensitive and pressure sensitive and which can be advantageously used according to the above-mentioned second embodiment are the recording material(s) described and claimed in the published Dutch Pat. application 67.04678.

In order to avoid a misunderstanding about the degree of hydrophobicity and hydrophilicity of the surface recording layer and interlayer or support respectively, it has to be understood that the surface layer in a sufficient degree has to be more hydrophobic than the underlying element in order to obtain a sufficient in water repellency differentiation for printing with a hydrophilic ink.

The hydrophobic surface recording layer may contain to a certain extent, e.g. from 1 to 80 percent by weight, dispersed solid substances which have a hydrophobic character, e.g. talcum, whereby the frangibility of the layer may be increased and the layer becomes better suited for pressure recording.

The hydrophilic interlayer may contain hydrophilic pigments, e.g. hydrated silicon dioxide, aluminum hydroxide, zinc oxide, barium sulfate, titanium dioxide, basic lead carbonate, barium carbonate, calcium carbonate, china clay and/or diatomaceous earth.

For enabling visual inspection of the obtained image differentiation in the recording material, the surface recording

layer may contain dye(s) or pigment(s) which can be removed together with the hydrophobic meltable or frangible substance of the recording layer. If a dark colored dye or pigment is used the heat sensitivity of the surface recording layer is substantially increased. The content of pigments and/or dyes may be up to 50 percent by weight with the proviso that the hydrophobicity is not substantially affected. Suitable pigments are, e.g., carbon black, graphite, oxides or sulfides of heavy metals, particularly of those heavy metals having an atomic weight between 45 and 210, such as manganese or lead sulfide or these heavy metals themselves in finely divided state such as silver, bismuth, lead, iron, cobalt or nickel. At present, preference is given to carbon black as heat-absorbing pigment.

These layers containing a large amount of light-absorbing pigment are preferably not used in a recording technique wherein the heat is conducted from the image markings to the recording layer but are used in a recording technique wherein the heat is generated in the recording material by an imagewise irradiation of the recording material with electromagnetic rays, e.g. visible light or infrared light modulated (transmitted or reflected) by a graphic original. The said irradiation is preferably a high intensity irradiation of short duration i.e. lasting no longer than 0.1 seconds. Such irradiation can be advantageously brought about by means of a flashlamp yielding a light energy of at least 0.1 w. sec. per cm.²

For such type of exposure and suited radiation absorbing pigments wherein heat is generated by absorption of electromagnetic radiation reference is made, e.g., to the published Dutch Pat. application 66.06719.

The support is preferably flexible to enable roller processing of the recording material and its bracing on the printing cylinder. Thus, the support may be a flexible metal foil, e.g. an aluminum foil, a paper sheet or resin film.

For the coating of the hydrophobic substances of the surface recording layer preferably a solvent is used which is not a solvent for the hydrophilic substances contained in the interlayer. Such solvent or solvent mixture is selected depending on the types of the components of both layers. Typical solvents for coating the hydrophobic substances are, e.g., hydrocarbons and halogenated hydrocarbons, including benzene, toluene, xylene, ligroin, trichloroethylene, carbon tetrachloride, n-hexane, methylene chloride and ethyl acetate. The hydrophobic layer may also be coated by sublimation or coated starting from melted hydrophobic substances with the proviso that during coating said substances do not penetrate in the hydrophilic interlayer or support. The coating of the layers may be carried out with conventional coating devices.

A type of hydrophilic printing ink which is particularly suited for use in reversed planographic printing is described and claimed in the United Kingdom Pat. application 7,800/65 (corresponding with Belgian Pat. specification 676,898), which specification and application should be read in conjunction herewith. Said ink is defined as a hydrophilic printing ink composition having dispersed therein a lipophilic phase which is colorless or substantially colorless or of a color tone contrasting with that of said printing ink.

An hydrophilic printing ink for use in reversed planographic printing is not necessarily mainly composed of water. In the colored hydrophilic phase hydrophilic compounds dissolved in water may be present in a relatively high amount, e.g. up to 80 percent by weight in respect of water, for conferring to the ink a higher viscosity and a more pastelike consistency. Hydrophilic compounds, which in that respect are particularly useful, are water-soluble polyols, e.g. ethylene glycol and water-soluble polyoxy-alkylene compounds. The hydrophilic phase preferably contains a wetting agent and may be colored by means of a water-soluble dye and/or a pigment. As pigment for black ink carbon black is preferred.

The colorless or substantially colorless hydrophobic phase of the printing ink (if such a phase is dispersed in the hydrophilic phase of used as "wetting liquid in a damping system for reversed planographic printing") preferably does not contain

a solvent for the hydrophobic substance(s) of the surface recording layer in order to avoid degrading of the printing master

One of the main advantages of the present invention resides in the completely dry production of a planographic printing master blank since if hydrophilization of the interlayer or support is necessary it may take place on the printing machine itself, e.g. during inking on revolving the printing cylinder before feeding copying paper.

The following example illustrates the present invention.

EXAMPLE

A paper support weighing 90 g. per m.² was coated at 45° C. pro rata of 36 m.² per with litre a composition comprising:

Water	ccs.	1, 000
Cyanamer P 26 (trade-name for a copoly(acrylamide/acid) sodium salt of acrylic acid) (80/4/16) made commercially available by American Cyanamid Co., New York, N. Y., U.S.A.)	g.	50
12.5% solution of saponine in a mixture of water and ethanol (80/20)	ccs.	10

The resulting dry layer was then covered at 60° C. with a wax layer pro rata of 36 sq.m per litre from a coating composition comprising:

Carbon tetrachloride	ccs.	1, 000
Warco wax 170 A yellow (trade-name for a microcrystalline saturated hydrocarbon wax with a melting range between 76 and 79° C. made commercially available by Warwick Wax Co., Inc., Long Island City, N. Y., U.S.A.)	g.	100

After drying, the wax layer side of the resulting recording material was placed in contact with an original, having infrared absorbing markings, the image markings facing the wax layer.

The composite element was then exposed reflectographically by means of an infrared radiation source facing the paper support of the recording material. A master ready for reversed planographic printing was obtained.

The master was then inked with a composition prepared by adding first whilst stirring

Urea	Grams	12. 5
1,6-hexane-diol		40
a 53% aqueous dispersion of copoly(n-butyl acrylate/vinyl acetate) (72/28)		20
80% aqueous solution of a melamineformaldehyde resin		37. 5

Triethanolamine	Grams	1. 5
30% aqueous solution of copoly(vinyl acetate/crotonic acid ammonium salt) (95/5)		110

to an amount of 250g. of an aqueous carbon dispersion comprising per 100g. 53g. of carbon, 23g. of water, 18g. of ethylene glycol, and 6g. of nonylphenylpolyethylene oxide. To the obtained mixture 300 cc. of white spirit (having a boiling range of 140° to 200° C.) were added and intimately dispersed therein with an ultrasonic wave generator.

The printing master braced on a printing cylinder was inked with the above described ink and legible copies were produced on common paper by direct-planographic printing, i.e. without offset roller.

The prepared printing master blank was also pressure sensitive so that by the impact of typewriter keys on the support a laterally reversed hydrophilic image was obtained. The "typed" printing master was suited for direct reversed planographic printing.

We claim:

1. A process for preparing planographic prints, comprising the steps of:

1. providing a heat-sensitive recording material comprising a support, a water-permeable hydrophilic colloid interlayer adjacent to said support and an external water-impermeable hydrophobic layer in contact with said colloid layer, said hydrophobic layer consisting essentially of at least one film-forming hydrophobic substance which melts on heating and in such molten condition is absorbed into said hydrophilic colloid layer;

2. subjecting said recording material to a pattern of heat to cause said hydrophobic substance in the heated regions to become absorbed into said water-permeable interlayer, thereby exposing portions of said hydrophilic layer in a pattern corresponding with the heat pattern;

3. inking the heated external layer with a hydrophilic printing ink; and

4. transferring said ink to a copy material to produce said print.

2. A process according to claim 1, wherein said hydrophobic substance is miscible in its molten state with the hydrophilic colloid of said interlayer.

3. A process according to claim 2, wherein the interlayer comprises a polyacrylamide and the surface recording layer mainly contains a paraffinic microcrystalline wax melting between 40 and 120°C.

4. A process according to claim 3, wherein the support is paper.

5. A process according to claim 1, wherein the printing ink consists of a colored hydrophilic continuous phase and a lipophilic phase distinct in color from said hydrophilic phase dispersed in said continuous phase.

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