HEXAMETHYLTRIAMINOPHENYLFLUORENE PRINTING COMPOSITION AND MANIFOLDING SHEET THEREWITH

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Patented Nov. 26, 1968

13 Claims. (Cl. 8—2.5)

ABSTRACT OF THE DISCLOSURE

The invention herein is the use of Fluorene Violet dye with the formula

![Formula Image]

where R is short chain alkyl group, in an ink or in their leuco form for oxidation to a colored form on paper or a fabric. The dye can also be applied as a pigment when reacted with molybdic or phosphotungstic acid. The 9-0 nitro derivative and oleic salt are produced and used in a manner similar to the dyes of the above structural formula.

This invention relates to a new series of light-stable printing compositions of high color intensity for use in printing operations that require intense colors. This invention further relates to coatings containing these printing compositions. This invention further embraces the superior recording forms that contain these novel printing compositions.

Since the discovery of Methyl Violet dye by Lauth in 1861 and its commercial manufacture in 1866, extensive experimental effort has been directed towards the utilization of basic arylmethene dyes in business printing systems. The reason for this is that the extremely high color intensity of arylmethene dyes such as Methyl Violet, Crystal Violet, and Victoria Blue permits the attainment of easily readable characters with a very small amount of dye. For this reason, the original hectograph system utilized Methyl Violet dye; and present-day spirit duplicators still use compositions containing Crystal Violet dye.

Before 1900, in Victorian times, the only method available for making copies of business letters and orders, other than to make a hand-written copy, was to write the original letter with a soft, solvent-soluble ink (usually containing Methyl Violet dye). This original was then run through a "letterpress" in contact with a wet sheet of paper to give a reverse copy of the original on the wet sheet. Needless to say, this wet treatment invariably caused bleeding of the original writing as well as the writing on the copy obtained; and such methods of avoiding hand copying of an original achieved only small-scale use.

Around 1900, the introduction of camoubla wax from Brazil into the United States almost immediately led to its use as a binder for the soft letterpress ink; and copy paper (today called "carbon paper") became a new item of commerce whereby it was now possible to make an exact copy of an original at the same time the original was made. The poor light-stability of the letterpress ink could not be overcome by the use of moldants; and this ink was eventually displaced by a carbon dope containing Methyl Violet toner (to give a true black print), which is still widely used today.

Since the introduction of carbon paper into business office procedure, the "average" number of copies required for a business communication has increased from none in 1900 to one in 1910 to anywhere from five through ten copies in 1960. Some office forms (such as railroad routing forms) today require that as many as fourteen copies be obtainable in one operation (such bulk forms now require the use of different printing compositions on different sheets; the top sheets have a very hard carbon dope backing and the bottom sheets have a very soft carbon dope backing). This is one major factor for the vast increase in the use of electric typewriters and ball point pens in industry; older writing and typing instruments simply cannot exert the heavy pressures that are necessary to make the large number of legible copies now required from the operation.

To make, handle, send, and receive this rapidly increasing number of office forms now requires the services of a very large staff of office workers in American industry. The labor costs involved in making, handling, and reading this large number of copies has become a rapidly increasing factor in business overhead. The great increase in research in the field of business printing operations during the past twenty-five years has resulted from the need not only to improve the properties of existing copy systems but also to develop new printing systems which are easier to prepare, use, and handle by inexperienced office help.

All systems of business recording represent a form of printing. Printing may be simply defined as the creation of indicia within or on something. Most forms of printing involve the making of indicia either by the transformation of a colorless ink coating onto a colorless substrate (in heat-sensitive copy papers) or by the transfer of an ink from one area of the printing system to the area where it forms colored indicia. This ink may be externally transferred from the reverse of one manifolding sheet (master) to the face of another sheet (receiving sheet or copy sheet) where it forms colored indicia; or the ink may be internally transferred from a protected (colorless) region within a sheet to reactive region where it forms color (as in several new colorless copy papers). It should be remembered that an ink is defined as a "fluid or viscous material, used for writing and printing"; the ink itself may be colored (as in ball point pen inks) or it may be colorless, as in sympathetic inks (which includes heat-sensitive inks, light-recording inks, etc.).

All of these inks, whether colored or colorless, when used in copy papers are held onto or within the surface of a base web (which may be paper, metal, or plastic) by "binders"; a binder may be considered simply as a "glue" to hold an ink onto a copy paper. The actual characteristics (color, light-stability, humidity resistance, etc.) of the print obtained depend solely upon the chemical nature of the ink used. Binders for printing compositions in business forms have been the subject of much recent research; and the original waxy binders used in copy papers for many years have recently been to some extent displaced by vinyl resins, by hydrophilic colloids, and by other nonwaxy materials.

At the present time the new copy papers recently brought onto the market use various arylmethene dye derivatives as the printing color; in other words, most of the successful "improvements" in business copy systems are primarily due to changes or improvements in the binders which hold the recording ink onto or within the recording form. The advantages conferred on the form by the use of a new binder are oftentimes diminished operations-wise by basic defects in the specific arylmethene color-former used; otherwise these new forms would rapidly displace standard carbon paper forms completely from the market. What has long been needed for
business printing operations is a new series of printing compositions of high color intensity, pleasing shade, and improved light-stability whose properties can be varied for applications in a wide variety of printing operations.

It is the purpose of this invention to provide improved printing compositions of high color intensity which are considerably more light-stable than present alylmethane printing compositions and which are considerably more adaptable for specialized formulation than are presently available printing compositions.

The printing compositions of the present invention make use of three different color forms of Fluorene Violet dyes: the intensely colored dye form (I), the colorless reduced (leuco) form (II), and the colorless basic (carbinol) form (III):

![Diagram of Fluorene Violet dyes]

\[
RN \quad NR_2 - OH \quad R_2N - \quad NR_2 \quad OH
\]

where R may be hydrogen and lower alkyl.

The colorless Fluorene Violet leucos (II), whose preparation is disclosed in my copending application, "Process for Converting Alylmethane Dyes Into Fluorene Dye Derivatives," Ser. No. 290,256, filed June 24, 1963, now Patent No. 3,544,189, are completely air-stable, being converted by strong oxidants or by activated surfaces into the intense dye (I). This dye can be "radicated" by strong reducing agents through conversion back to the stable leuco; this is not practiced in commercial operations with the easily reoxidized alylmethane leucos, such as leuco Crystal Violet. As the Fluorene Violet dye bases (III) are less basic than Crystal Violet base and form more spirit-soluble colors, their coated compositions are not as staining to handle as the Crystal Violet compositions.

The spirit-soluble feature of Pentamethyl Fluorene Violet salts and Hexamethyl Fluorene Violet salts makes their use in spirit duplicator inks and ball point pen inks of particular merit. The prints made on certain textiles, particularly woolens and nylon, are more resistant to washing and are more light-stable than the corresponding prints from Methyl Violet dye. Although the chloride and acetate salts of the alkylated Fluorene Violet dyes are alcohol-soluble, in order to achieve high solubility in commercial nonaqueous ink vehicles it is oftentimes desirable to convert the dye bases into the salts of water-insoluble carboxylic acids containing more than six carbon atoms. For this purpose, oleic acid, stearic acid, ricinoleic acid, montanic acid and other wax acids, abietic acid and other resin acids, lauric acid, etc., are particularly suitable. These ink vehicle-soluble salts are readily prepared in situ by adding the finely powdered dye base to a warm solution or suspension of the carboxylic acid in the desired ink vehicle or to the liquid form of the carboxylic acid. For example, Pentamethyl Fluorene Violet base added to warm oleic acid forms the intensely colored oleate salt.

While the Fluorene Violet dye derivatives used in the compositions of the present invention are considerably more light-stable than the paraarsaniline alylmethane dye colors, like all organic dyes they will eventually fade in strong sunlight due to the thermodynamic transformation whereby all things organic exposed to oxygen inevitably pass away:

\[
C_6H_4O_2 + O_2 \rightarrow C_6O_2 + H_2O
\]

The dye-containing printing compositions of the present invention represent a marked improvement over the alylmethane colors and they will last indefinitely under normal conditions of use; but they are not in the range of the few so-called permanent pigments.

Systems of extreme light-stability, however, are obtained by the use of phosphomolybdate and phosphotungstate toners of the Fluorene Violet dyes. These new toners give printing compositions of extremely deep shade. The toners from Hexamethyl Fluorene Violet, for example, are a deep blue-black in natural sunlight and appear black under artificial incandescent light. The toners from Mono Nitro Hexamethyl Fluorene Violet are a true black in sunlight. These toners are more light-stable than the Methyl Violet toners used in present printing compositions; and colored films obtained through the use of these new Fluorene Violet toners hold up well under exposure to sunlight. Their deep shades, combined with extremely good light-stability, makes these toners of value in films and coatings where their low solubility prevents migration and "bleeding."

The color shade obtained by the present printing compositions depends primarily upon the dye concentrations used. A very low dye concentration gives a bright, slightly fluorescent violet; but this very slight fluorescence is readily quenched by impurities and by higher dye concentrations. The usual color obtained is indigo to deep violet, being indigo at usual concentrations. The toners yield a deep blue-black to black shade. The most intense black shades are obtained with the use of the black pigments from alkylated Mononitro Fluorene Violet dyes and phosphomolybdic acid or phosphotungstic acid.

The printing compositions of the present invention are subject to considerable formula variation, using an extensive selection of binders, solvents, and stabilizers, such as chromic, iron, and copper salts. In the printing ink industry an ink formulation is like a cookbook recipe: merely a starting point for individual variation. The formula of any given commercial ink composition is greatly dependent upon economic factors such as costs and availability of materials involved; a change in the price of any material in an Ink composition by 1/4¢ to 1/2¢ a pound will immediately lead to extensive reformulation of the entire ink composition. For this reason, considerable variation in the examples given will be the rule rather than the exception; and the invention should not be considered limited to the examples cited.

**DESCRIPTION**

The starting materials were prepared as disclosed in my copending application, "Process for Converting Alylmethane Dyes Into Fluorene Dye Derivatives," Ser. No. 290,256, filed June 24, 1963. A typical preparation of Hexamethyl Fluorene Violet Base (2,7-bis(dimethylaminomethylene) -9-p-carboxyfluorenone-o)-9) was as follows: 94.7 grams of Hexamethyl Fluorene Violet base was dissolved in warm glacial acetic acid, and 62.5 grams of dry lead peroxide was added. Considerable heat is evolved. The solution was allowed to stand for one hour, then water and sulfuric acid was added to precipitate out the lead as lead sulfate. After four hours, the lead sulfate was filtered off, and the cold filtrate was made alkaline to precipitate out the Hexamethyl Fluorene Violet base. The dye base was filtered off and washed well with water.
5 containing some ammonia. The yield of dried pale violet powder is 90-95%. The mononitro derivatives of the Fluorene Violet dyes are easily prepared by nitration of the dye dissolved in cold concentrated sulfuric acid with the calculated amount of nitric acid. A typical preparation was as follows: 45 grams of Hexamethyl Fluorene Violet base was dissolved in 400 grams of warm concentrated sulfuric acid, and the solution was cooled to 3°C. A cold solution of 7 ml. of concentrated nitric acid in 40 ml. of concentrated sulfuric acid was added in portions, maintaining the reaction temperature below 15°C. The solution was allowed to stand at room temperature for five hours, then poured onto ice. The cold solution was then made alkaline, and Mononitro Hexamethyl Fluorene Violet base (2,7-bis(dimethylamino)-9-o-nitro-p-dimethylaminophenylfluorene - ol - 9) precipitated out. The dye base was filtered off and washed with water. The yield of pale green powder is about 95%. The pure material is yellow and the solutions in toluene and acetone are yellow. This is a weak dye base which does not stain skin. There is considerable steric hindrance at the central carbon atom of these nitratated fluorene dye bases, which inhibits attainment of planarity.

To prepare salts of the dyes with organic acids, one merely adds the finely powdered dye base to the liquid form of the acid or to the solution or dispersion of the acid in the desired ink vehicle.

**Example 1**

Five grams of Pentamethyl Fluorene Violet base were added to five grams of warm oleic acid to give an intensely colored paste of Pentamethyl Fluorene Violet oleate. To this was added a hot (90°C.) 15% molten composition of twenty-five grams of oricury wax, ten grams of paraffin wax, and forty grams of petrolatum. The resultant hot dope was coated at 85°C. onto paper by standard coating techniques to give a one-time copy paper which yielded an intense indigo print on a paper receiving sheet.

**Example 2**

Five grams of Hexamethyl Fluorene Violet base was added to a warm methyl ethyl ketone solution of five grams of abietic acid rosin) and three grams of tributyl phosphate to form a solution of Hexamethyl Fluorene Violet abietinate. When this ink was used as a marking fluid on metal surfaces evaporation of the solvent gave a deep blue print which was resistant to removal by water and by rubbing.

**Example 3**

Four grams of Hexaethyl Fluorene Violet base (2,7-bis(diethylamino) - 9-p-diethylaminophenylfluorene-ol-9) were added to a hot (85°C.) melt composed of twenty-five grams of montan wax containing montanic acid) and thirty grams of petrolatum to form a solution of Hexaethyl Fluorene Violet montanate in the hot dope. This was coated onto paper by standard coating techniques to give a one-time copy paper, which gave intense indigo indicia on a paper receiving sheet.

**Example 4**

Five grams of Mononitro Hexamethyl Fluorene Violet base was dissolved in acetone, and to this was added an acetone solution of seven grams of phosphomolybdic acid. An intense black powder precipitated out. The acetone was removed. The black powder was dispersed in linseed oil to form a deep black ink with excellent hiding power and light-stability. This gave deep black prints from an inked metal plate.

**Example 5**

Five grams of Mononitro Pentamethyl Fluorene Violet base was dissolved in methyl ethyl ketone, and a solution of seven grams of phosphotungstic acid in acetone was added. An intense black powder precipitated out, and the solvent was removed. The black pigment was dispersed in a mixture of castor oil and oleic acid to form a black ink. This was used to ink a typewriter ribbon which gave intense black prints on a receiving sheet.

**Example 6**

Five grams of Pentamethyl Fluorene Violet base were dissolved in n-propanol and eight grams of phosphomolybdic acid in acetone were added. The deep blue-black powder which formed, after removal of the acetone, appeared black under artificial incandescent light but deep blue-black under strong sunlight. The pigment was dispersed in oleic acid to form an ink which yielded almost black prints on paper.

**Example 7**

Five grams of Hexaethyl Fluorene Violet base was dissolved in ethanol, and a solution of six grams of phosphotungstic acid in ethanol was added. The deep blue-black powder which formed was, after removal of the ethanol, dispersed in polyethylene glycol to give an almost black writing fluid for stylus instruments.

**Example 8**

Five grams of Pentamethyl Fluorene Violet base was dissolved in acetone, and to this was added an acetone solution containing four grams of phosphomolybdic acid and four grams of phosphotungstic acid to form an almost black precipitate. After removal of the acetone, the dry precipitate was added to a hot (85-90°C.) melt of twenty grams of carnauba wax and forty grams of petrolatum; and the hot black dope was coated onto paper by standard coating techniques to give a one-time copy paper. This gave black indicia on a paper receiving sheet.

Colorless printing compositions and colorless coatings may be obtained by the use of Fluorene Violet leuco, which are colorless and nonstaining to skin. These leucos form colorless salts with strong acids; these salts are soluble in water, glycols, and other highly polar solvents. Unlike the porosaniline leucos, such as Crystal Violet Leuco, the colorless solutions of the Fluorene Violet leucos are not oxidized by air in the presence of an activator; for this reason, they do not undergo premature coloration when used in inert coatings.

**Example 9**

2.5 grams of Hexamethyl Fluorene Violet leuco were dissolved in 98 grams of chlorinated diphenyl to form a substantially colorless solution. This gave intense indigo prints on a chloranil-coated paper. This solution was emulsified in an aqueous polyvinyl alcohol solution; and this emulsion was coated onto paper by standard coating techniques to give a colorless coating. The air-dried sheet was used to print on a chloranil-coated paper to give intense violet prints.
Three grams of Hexaethyl Fluorene Violet leuco were dissolved in 97 grams of diethylbenzene to form a substantially colorless solution. This was emulsified in warm gelatin solution made basic by the addition of a small amount of sodium carbonate solution, and the emulsion was coated onto paper by standard coating techniques to give a colorless coating which yielded intense prints on a chlorinil-coated paper.

Example 11

Three grams of Hexamethyl Fluorene Violet leuco were dispersed in water, and enough toluenesulfonic acid was added to form the water-soluble toluenesulfonic acid salt of the leuco. The water was removed to give a white crystalline solid which was dissolved in 100 grams of polyethylene glycol to form a colorless, nonstaining solution. When this was used to print on freshly prepared sodium dichromate-coated paper, intense indigo indicia were obtained which suffered almost no fading after three months exposure under glass to sunlight.

Example 12

Five grams of Hexamethyl Fluorene Violet leuco were dispersed in water, and enough dilute sulfuric acid was added to form the water-soluble sulfate salt. This was used to print intense indigo indicia on a sodium dichromate paper; the resultant prints showed almost no fading after exposure for four months under glass to sunlight. The colorless solution of the sulfate salt may also be used to print colorless designs on textiles. Immersion of the cloth in a sodium dichromate bath then gives intense deep violet to indigo prints on the textile.

The substantially colorless Fluorene Violet dye bases give colorless solutions in low polar solvents; these colorless solutions react with acids to form intensely colored salts. The colorless recording solutions thus obtained are not as reactive as those containing Crystal Violet base, but they react with paper fibers (cellulose) to give very light-stable colors which are very resistant to removal or bleeding by water washing.

Example 13

Three grams of Hexamethyl Fluorene Violet base were dissolved in 100 grams of diethylbenzene to form a substantially colorless solution. This recording solution gives intense indigo prints on clay-coated papers.

Example 14

Four grams of Hexaethyl Fluorene Violet base were dissolved in 105 grams of chlorinated diphenyl to give a substantially colorless recording solution which gave intense indigo prints on clay-coated papers.

Example 15

Three grams of Mononitro Hexamethyl Fluorene Violet base were dissolved in 100 grams of methyl ethyl ketone to give a yellow solution which did not stain skin. This was used to print on a receiving sheet impregnated with phosphomolybdic acid to give an intense black print.

Aqueous colored printing fluids using the water-soluble colored salts of Fluorene Violet dyes give very light-stable prints on cellulose (paper fibers), apparently because of the formation of some dye-fiber combination. Prints obtained by the use of aqueous Fluorene Violet dye solutions exhibit considerable resistance to water-bleeding, unlike the prints obtained by the use of water-soluble aroylmethane dye salts such as Crystal Violet dye on paper which bleed badly upon addition of water. This water-resistance exhibited by aqueous Fluorene Violet dye solutions after drying on fibers makes these aqueous printing compositions of value in water-base writing and printing fluids for paper and textiles.

Example 16

Approximately 0.75 gram of pure Hexamethyl Fluorene Violet acetate is dissolved in a solution of 97 grams of water and 3 grams of glacial acetic acid to form an indigo-colored solution. A fountain pen filled with this writing fluid gave intense indigo prints of excellent readability on bond paper. These prints showed only slight fading after three months exposure under glass to sunlight.

It should be clearly understood that a wide variety of solvents, ink resins, and binders can be used to formulate the printing compositions of the present invention; and the invention should not be considered limited to the examples cited. Additives to control adhesion to surfaces, to change wetting properties, to control solvent volatility and other minor properties may be added for specialized applications. The fluids, coatings, and forms containing the unusually pure Fluorene Violet derivatives made available by an unexpected discovery by the author of the present invention offer superior printing compositions for use in many diversified printing and coloring applications.

Having described my invention, I claim:

1. A printing medium comprising the intensely colored fluorene dye represented by the following formula:

   ![Formula Image]

   where R is chosen from the group consisting of hydrogen and lower alkyl, said dye dissolved in a solvent therefor, said solvent dispersed in an organic binder to form an intensely colored recording ink.

2. A printing medium comprising the fluorene dye base represented by the following formula:

   ![Formula Image]

   where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefore, said solvent dispersed in an organic binder to form a recording ink which is structurally adapted to form intensely colored indicia upon contact with acid surfaces.

3. A printing medium comprising the fluorene dye leuco represented by the following formula:

   ![Formula Image]

   where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefor, said solvent dispersed in an organic binder to form a recording ink which is structurally adapted to form
intensely colored indicia upon contact with a strong oxidant.

4. A water-base printing medium comprising the intensely colored fluorene dye represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dispersed in an aqueous vehicle, said aqueous vehicle dispersed in a hydrophilic colloid to form an intensely colored recording ink.

5. The method of printing which comprises applying to a receptive surface an intensely colored ink which comprises a fluorene dye represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dispersed in an organic binder.

6. The method of printing which comprises applying to an acid surface a substantially colorless liquid which comprises a fluorene dye base represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefore, said compound being structurally adapted to react with the acid surface to form an intensely colored dye.

7. The method of printing which comprises applying to an oxidizing surface a substantially colorless liquid which comprises a fluorene dye leuco represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefore, said compound being structurally adapted to form intensely colored indicia upon contact with a strong oxidant.

8. A printing composition comprising the substantially colorless fluorene dye leuco represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a hydrophilic colloid to form a substantially colorless recording medium.

9. A printing composition comprising the salt of a strong acid with the substantially colorless fluorene dye leuco represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said salt dissolved in a solvent therefore, said compound being structurally adapted to form intensely colored indicia upon contact with a strong oxidant.

10. A printing composition having as a constituent thereof a substantially colorless fluorene dye leuco represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefore, said compound being structurally adapted to form intensely colored indicia upon contact with a strong oxidant.

11. A manifolding sheet comprising a web having a coating on one side thereof, said coating comprising a solvent and the intensely colored fluorene dye represented by the following formula:

where R is chosen from the group consisting of hydrogen and lower alkyl, said compound dissolved in a solvent therefore, said compound being structurally adapted to form intensely colored indicia upon contact with a strong oxidant.
where R is chosen from the group consisting of hydrogen and lower alkyl dispersed in said solvent.

12. A manifolding sheet comprising a web having a coating on one side thereof, said coating comprising a solvent and the substantially colorless fluorene dye leuco represented by the following formula:

\[
\begin{align*}
 & \text{RN} - \text{N} - \text{NR}_2 \\
 & \text{NR}_3
\end{align*}
\]

where R is chosen from the group consisting of hydrogen and lower alkyl dispersed in said solvent.

13. A manifolding sheet comprising a web having a coating on one side thereof, said coating comprising a solvent and the substantially colorless fluorene dye base represented by the following formula:

\[
\begin{align*}
 & \text{RN} - \text{OH} - \text{NR}_2
\end{align*}
\]

where R is chosen from the group consisting of hydrogen and lower alkyl dispersed in said solvent.

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