

[54] **PRESSURE-SENSITIVE COPYING PAPER  
CONTAINING PHENOXAZINE  
COMPOUNDS**

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[51] Int. Cl. .... **B41m 5/16**

[58] Field of Search ..... **117/36.2, 36.8; 260/243 A, 260/244 R**

[56] **References Cited**

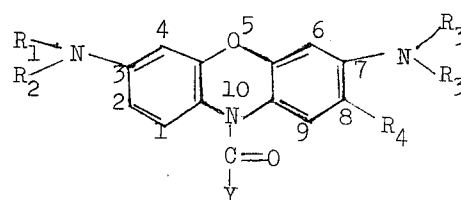
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[57] **ABSTRACT**

A pressure-sensitive copying paper comprising an adsorbent solid acid and a microencapsulated color former capable of forming a distinct color when reacted with the adsorbent acid coated on the same or a different surface of a support or supports, the microencapsulated color former being at least one phenoxazine compound represented by the formula



wherein R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub> and Y are as defined hereinafter, is disclosed.

**2 Claims, No Drawings**

# PRESSURE-SENSITIVE COPYING PAPER CONTAINING PHENOXAZINE COMPOUNDS

## BACKGROUND OF THE INVENTION

### 1. Field of the Invention

The present invention relates to a pressure-sensitive copying paper and, more particularly, to a pressure-sensitive copying paper having a microcapsule layer containing a novel phenoxazine compound as a color former.

### 2. Description of the Prior Art

Pressure-sensitive copying papers generally comprise a transfer sheet having thereon a layer of fine capsules containing therein a solution of an electron donating colorless organic compound (hereinafter referred to as "color former") in an organic solvent, and a receiving sheet having thereon a layer of an electron accepting solid. A suitable binder is generally used for coating the solid.

When both sheets are put together in such a manner that an electron donating layer is brought into contact with an electron accepting layer and localized pressure is applied to the combination of sheets by handwriting or typewriting, the capsules are ruptured and the colorless color former contained in the capsules is adsorbed by the electron accepting solid, which is usually a solid acid, to form a color.

Pressure-sensitive copying paper systems comprising the aforesaid transfer sheet (hereinafter referred to as "upper sheet") and a receiving sheet (hereinafter referred to as "lower sheet") and an intermediate sheet (hereinafter referred to as "middle sheet") are also known. In these systems the middle sheet is usually coated, on opposite surfaces, with a layer of microcapsules containing a color former solution and with a layer of a solid acid and a binder, respectively.

Another type of pressure-sensitive copying system is a recording sheet which can be prepared by applying the above described microcapsules and the adsorbent solid acid on the same surface of a support.

The solid acid used in the conventional type of pressure-sensitive copying papers includes active clay substances such as acid clay, atapalgite, zeolite, bentonite and the like, solid organic acids such as succinic acid, tannic acid, benzoic acid and the like, and acidic organic polymers such as phenol-formaldehyde polycondensates, phenol-acetylene copolymers, styrene-maleic anhydride copolymers containing residual acidic radicals, salicylic acid-formaldehyde polycondensates and the like. The organic solvent for dissolving the color former which is conventionally used includes, for example, ethylene glycol, chlorobenzenes and chlorobiphenyls.

Benzoyl leuco methylene blue is well known as a color former forming a blue color. Benzoyl leuco methylene blue slowly forms a blue color on a lower sheet when an upper sheet having the benzoyl leuco methylene blue thereon is superposed on a lower sheet coated with an active clay substance such as acid clay and localized pressure is applied to the assembly by, e.g., handwriting. However, this compound does not form a clear blue color when a lower sheet coated with an acidic organic polymer such as a phenol-formaldehyde polycondensate is used. It has, therefore, strongly been desired to develop pressure-sensitive copying papers which are capable of forming a blue color with an excellent fastness to light even on a lower sheet coated

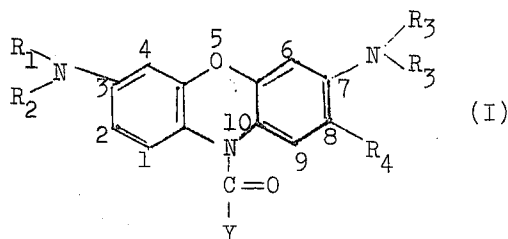
with the above described acidic organic polymer, because such pressure-sensitive copying papers are expected to be useful in a wide variety of utilities.

An object of the present invention is, therefore, to provide pressure-sensitive copying papers containing as a color former a phenoxazine compound which rapidly forms a stable blue color having excellent light fastness and water resistance when it is brought into contact with a solid acid.

Another object of the present invention is to provide pressure-sensitive copying papers which can be colored various colors by using a phenoxazine compound in combination with other known color formers.

## SUMMARY OF THE INVENTION

According to the present invention, there is provided a pressure-sensitive copying paper containing as a color former a phenoxazine compound represented by the formula



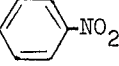
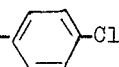
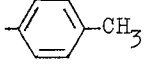
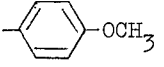
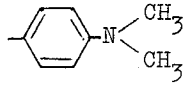
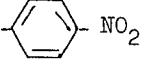
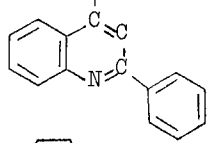
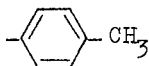
wherein R<sub>1</sub> and R<sub>3</sub> each represents a lower alkyl group; R<sub>2</sub> represents a lower alkyl or a phenyl group; R<sub>4</sub> represents a hydrogen atom or a methyl group; and Y is selected from the group consisting of alkyl, alkenyl, styryl, benzyl, phenoxymethylene, phenyl, naphthyl, furyl, thienyl, pyridyl and quinolyl groups in which the styryl, benzyl, phenoxymethylene, phenyl and naphthyl groups may be substituted in the nucleus thereof by one of a lower alkyl, a lower alkoxy, a di-lower-alkylamino, a hydroxy, a chloro or a nitro group and the quinolyl group may be substituted in 2-position by either of a lower alkyl or a phenyl group.

## DETAILED DESCRIPTION OF THE INVENTION

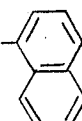
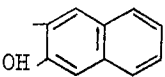
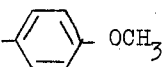
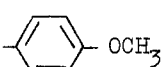
As the result of detailed investigations on color formers capable of forming a blue color on a lower sheet coated with an acidic organic polymer as a solid acid, it was found that, a blue color having an excellent light fastness is gradually formed on a lower sheet when localized pressure is applied by, for example, handwriting to an upper sheet coated with the phenoxazine compound represented by the above formula (I) as a color former superposed on a lower paper coated with an acidic organic polymer, active clay substance or a mixture thereof as a solid acid. It was also found that any desirable color having an excellent light fastness can be formed on a lower sheet when localized pressure is applied by, for example, handwriting to an upper sheet coated with the phenoxazine compound of the above formula (I) in combination with a known color former as a color former superposed on a lower sheet coated with an acidic organic polymer, active clay substance or a mixture thereof as a solid acid.

Representative examples of the phenoxazine compounds of the formula (I) which are particularly useful in the pressure-sensitive copying paper of the present

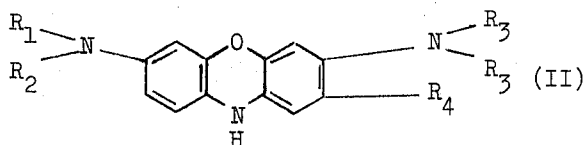
invention and the processes for the synthesis of the compounds are illustrated hereinafter.

| Color  | Substituents |                               |                               |                               |                                                                                                                         | Chemical Name                                                             |
|--------|--------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------|
|        | Former       | R <sub>1</sub>                | R <sub>2</sub>                | R <sub>3</sub>                | R <sub>4</sub> Y                                                                                                        |                                                                           |
| No. 1  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H -CH <sub>3</sub>                                                                                                      | 3,7-bis(Diethylamino)-10-acetylphenoxazine                                |
| No. 2  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H -CH=CH-CH <sub>3</sub>                                                                                                | 3,7-bis(Diethylamino)-10-crotonoylphenoxazine                             |
| No. 3  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H -CH=CH-                              | 3,7-bis(Diethylamino)-10-cinnamoylphenoxazine                             |
| No. 4  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H -CH <sub>2</sub> -  -NO <sub>2</sub> | 3,7-bis(Diethylamino)-10-(p-nitrophenylacetyl)-phenoxazine                |
| No. 5  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H -CH <sub>2</sub> -O-  -Cl            | 3,7-bis(Diethylamino)-10-(p-chlorophenoxyacetyl)-phenoxazine              |
| No. 6  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                      | 3,7-bis(Diethylamino)-10-benzoylphenoxazine                               |
| No. 7  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H  -CH <sub>3</sub>                    | 3,7-bis(Diethylamino)-10-(p-methylbenzoyl)-phenoxazine                    |
| No. 8  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H  -OCH <sub>3</sub>                  | 3,7-bis(Diethylamino)-10-(p-methoxybenzoyl)-phenoxazine                   |
| No. 9  |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H  -N(CH <sub>3</sub> ) <sub>2</sub> | 3,7-bis(Diethylamino)-10-(p-dimethylaminobenzoyl)-phenoxazine             |
| No. 10 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H OH                                 | 3,7-bis(Diethylamino)-10-(o-hydroxybenzoyl)-phenoxazine                   |
| No. 11 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H  -NO <sub>2</sub>                  | 3,7-bis(Diethylamino)-10-(p-nitrobenzoyl)-phenoxazine                     |
| No. 12 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                    | 3,7-bis(Diethylamino)-10-(2-furoyl)-phenoxazine                           |
| No. 13 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                    | 3,7-bis(Dimethylamino)-10-(2-thienoyl)-phenoxazine                        |
| No. 14 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                    | 3,7-bis(Diethylamino)-10-nicotinoylphenoxazine                            |
| No. 15 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                    | 3,7-bis(Diethylamino)-10-isonicotinoylphenoxazine                         |
| No. 16 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | H                                    | 3,7-bis(Diethylamino)-10-(2-phenylcinchonoyl)-phenoxazine                 |
| No. 17 |              | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub> | CH <sub>3</sub>  -CH <sub>3</sub>    | 3-Diethylamino-7-dimethyl-amino-8-methyl-10-(p-methylbenzoyl)-phenoxazine |

- Continued

| Color Former | Substituents                  |                                                                                   |                                 |                 | Y                                                                                 | Chemical Name                                                                  |
|--------------|-------------------------------|-----------------------------------------------------------------------------------|---------------------------------|-----------------|-----------------------------------------------------------------------------------|--------------------------------------------------------------------------------|
|              | R <sub>1</sub>                | R <sub>2</sub>                                                                    | R <sub>3</sub>                  | R <sub>4</sub>  |                                                                                   |                                                                                |
| No. 18       | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub>                                                     | CH <sub>3</sub>                 | CH <sub>3</sub> |  | 3-Diethylamino-7-dimethylamino-8-methyl-10-(1-naphthoyl)-phenoxazine           |
| No. 19       | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub>                                                     | CH <sub>3</sub>                 | CH <sub>3</sub> |  | 3-Diethylamino-7-dimethylamino-8-methyl-10-(3-hydroxy-2-naphthoyl)-phenoxazine |
| No. 20       | C <sub>2</sub> H <sub>5</sub> | C <sub>2</sub> H <sub>5</sub>                                                     | n-C <sub>4</sub> H <sub>9</sub> | H               |  | 3-Diethylamino-7-di-n-butylamino-10-(p-methoxybenzoyl)-phenoxazine             |
| No. 21       | CH <sub>3</sub>               |  | C <sub>2</sub> H <sub>5</sub>   | H               |  | 3-(N-Methyl-N-phenylamino)-7-diethylamino-10-(p-methoxybenzoyl)-phenoxazine    |

The compounds represented by the above formula (I) can be prepared by reacting 1 mole of a leuco compound represented by the formula (II)



wherein R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub> and R<sub>4</sub> are as defined above with respect to the formula (I), with 1.0 to 5.0 moles of an acid halide represented by the formula (III).



wherein X is a halogen atom and Y is as defined above with respect to the formula (I), in the presence of 1.0 to 10.0 moles of an acid acceptor such as sodium hydroxide, sodium carbonate, pyridine or triethylamine in a volatile organic inert solvent such as chloroform, benzene, toluene or chlorobenzenes or a mixture of such a volatile organic inert solvent and water, optionally while passing gaseous nitrogen through the reaction mixture, at a reaction temperature of from 30° to 130°C for a period of from 1 to 5 hours. After completion of the reaction, the organic solvent layer is separated, and is treated with a 3 to 10% by weight aqueous hydrogen peroxide solution, washed with water and distilled under reduced pressure to remove any residual organic solvent to obtain the desired color former having the formula (I). If necessary, the color former thus obtained can further be purified, for example, by recrystallization.

The preparation of the individual color formers as listed in the previous table are illustrated by the following production examples in which all parts given are by weight unless otherwise indicated.

#### PRODUCTION EXAMPLE 1

##### Preparation of Color Former No. 1

A mixture of 2.8 parts of 3,7-bis(diethylamino)-phenoxazine, 2.4 parts of acetyl chloride and 3.4 parts

of pyridine was heated at a reaction temperature of from 50° to 60°C for 2 hours in 100 parts of chloroform. Thereafter the mixture was treated with a 3% aqueous hydrogen peroxide solution and thoroughly washed with water. The chloroform layer was then separated and treated with an activated carbon, distilled under reduced pressure to remove the residual chloroform and recrystallized from ethyl alcohol to obtain 2.5 parts of a light brown-colored compound having a melting point of 146°-147°C. A solution of the resulting compound in benzene slowly turned blue when brought into contact with an acid clay or a phenolformaldehyde polycondensate coated on a paper.

The same color former was also obtained by repeating the above procedure using acetyl bromide in place of acetyl chloride.

#### PRODUCTION EXAMPLE 2

##### Preparation of Color Former No. 6

To a mixture of 60 parts of toluene and 50 parts of water were added 3.2 parts of 3,7-bis(diethylamino)-phenoxazine and 4.8 parts of benzoyl chloride, followed by the addition of 14.5 parts of a 17% aqueous sodium hydroxide solution. The mixture was then heated at a reaction temperature of from 50° to 55°C for 3 hours and thereafter worked-up in the same manner as described in Production Example 1 to obtain 3.7 parts of a light yellow-colored color former having a melting point of 123°-125°C. A solution of the resulting color former in benzene slowly turned blue when brought into contact with an acid clay or a phenolformaldehyde condensate coated on a paper.

#### PRODUCTION EXAMPLE 3

##### Preparation of Color Former No. 21

To a mixture of 100 parts of toluene and 80 parts of water were added 3.6 parts of 3-(N-phenyl-N-methylamino)-7-diethylamino-phenoxazine and 6.8 parts of p-methoxybenzoyl chloride, followed by the addition of 14.5 parts of a 17% aqueous sodium hydroxide solution. The mixture was then heated at a re-

action temperature of from 55° to 60°C for 2 hours and thereafter worked-up in the same manner as described in Production Example 1 to obtain 4.0 parts of a light yellow-colored compound having a melting point of 141°-143°C. A solution of the resulting compound in benzene slowly turned blue upon contact with an acid clay or a phenolformaldehyde condensate coated on a paper.

### PRODUCTION EXAMPLE 4

Preparation of Color Former Nos. 2 to 5, 7 to 13 and 17 to 20

Each of the Color Formers Nos. 2 to 5, 7 to 13 and 17 to 20 was prepared in a similar manner as described in Production Example 2 but using appropriate starting compounds of the formulae (II) and (III). The color formers thus obtained were slightly colored crystals having melting points and an appearance as shown in the following table.

| Color Former | Melting Point (°C) | Appearance of Crystals       |
|--------------|--------------------|------------------------------|
| No.2         | 110 - 112          | Light brown powder           |
| No.3         | 134 - 135          | Light yellow powder          |
| No.4         | 134 - 136          | Light brown powder           |
| No.5         | 125.5-127          | Pale white powder            |
| No.7         | 122 - 124          | Light yellowish brown powder |
| No.8         | 124.5-126          | Light yellow powder          |
| No.9         | 162 - 163          | Pale white powder            |
| No.10        | 85 - 90            | Light green powder           |
| No.11        | 142 - 143          | Light brown powder           |
| No.12        | 119 - 120          | Light yellowish green powder |
| No.13        | 163 - 166          | Light yellow powder          |
| No.17        | 121 - 123          | Light yellow powder          |
| No.18        | 103 - 105          | Light yellowish green powder |
| No.19        | 103 - 107          | Light green powder           |
| No.20        | 92 - 93            | Light yellow powder          |

### PRODUCTION EXAMPLE 5

Preparation of Color Former Nos. 14 to 16

To a mixture of 150 parts of toluene and 10 parts of triethylamine were added under a nitrogen stream 3.2 parts of 3,7-bis(diethylamino)-phenoxazine and the corresponding acid halide, i.e., 5.3 parts of nicotinoyl chloride for Color Former No. 14, 5.3 parts of isonicotinoyl chloride for Color Former No. 15, or 9.8 parts of 2-phenylcinchonoyl chloride for Color Former No. 16. The reaction mixtures in each instance was heated at a reaction temperature of from 50° to 55°C for 2 hours under a nitrogen stream and thereafter worked-up in the same manner as described in Production Example 1 to obtain the desired color formers, i.e., 3.8 parts of Color Former No. 14, 3.0 parts of Color Former No. 15 or 4.2 parts of Color Former No. 16, as slightly colored crystals in each instance. The melting points and appearance of the color formers thus obtained are shown in table below.

| Color Former | Melting Point (°C) | Appearance of Crystals       |
|--------------|--------------------|------------------------------|
| No. 14       | 139 - 141          | Light yellowish green powder |
| No. 15       | 117 - 118          | Light green powder           |
| No. 16       | 183 - 185          | Light yellow powder          |

In preparing pressure-sensitive copying papers using the phenoxazine compounds represented by the formula (I) as color formers, the well-known process disclosed in U.S. Pat. Nos. 2,800,457 and 2,800,458, i.e. a process which utilizes the phenomenon of complex coacervation can advantageously be employed. In this process, the color former is generally used in the amount of from 0.5 to 5.0% by weight relative to the amount of an organic solvent such as ethylene glycol, chlorobenzene or chlorobiphenyl as previously described.

The preparation of pressure-sensitive copying papers using some of the color formers represented by the formula (I) is illustrated in the following examples, in which all parts given are by weight unless otherwise indicated.

### EXAMPLE 1

Preparation of Pressure-sensitive Copying Paper

Two parts of Color Former No. 21 was dissolved in 100 parts of trichlorobiphenyl and to the solution were added 20 parts of gum arabic and 160 parts of water while stirring at 50°C to form an emulsion. To the emulsion were added, with stirring 20 parts of an acid treated gelatin and 160 parts of water followed by the addition of acetic acid to adjust the pH to 5. 500 parts of water was then added to the emulsion to effect the coacervation thereby forming a dense liquid film of gelatin-gum arabic around the surface of each droplet of trichlorobiphenyl having the color former dissolved therein and, after adjustment of the pH of the mixture to 4.4, 3.8 parts of 37% formalin was added thereto to harden the liquid film. The emulsion was then cooled to 10°C and, after adjustment of the pH to 9 by addition of an aqueous sodium hydroxide solution, allowed to stand to complete the encapsulation process. The liquid containing the microcapsules was applied using, for example, a roll coating or air-knife coating procedure to a paper and dried to obtain a colorless coated paper, i.e., an upper sheet. When the upper sheet thus obtained was superposed on a lower sheet coated with, as a solid acid, an acidic organic polymer, active clay substance or a mixture thereof in such a manner that the layer of microcapsules was brought into contact with a layer of the above solid acid and localized pressure was applied to the assembly of sheets by e.g., handwriting, a blue color was gradually formed in the pressed areas of the lower sheet. This blue color changed to a deep shade within several hours. No substantial discoloration of the completely developed blue color was observed when the lower sheet was exposed directly to sun light for a long period of time.

### EXAMPLE 2

Preparation of Pressure-sensitive Copying Papers

2.0 parts of each of Color Former Nos. 1 to 20 were separately processed in the same manner as described in Example 1 to form separate liquids containing microcapsules. The liquid was then applied to a paper and dried to obtain a colorless upper sheet. When the upper sheet was superposed on a lower sheet coated with, as a solid acid, an acidic organic polymer, active clay substance or a mixture thereof and localized pressure was applied to the assembly of sheets by, e.g., handwriting, a blue color was formed immediately in the pressed areas of the lower sheet. No substantial discoloration of

the blue color was observed when the lower sheet was exposed directly to sun light for a long period of time.

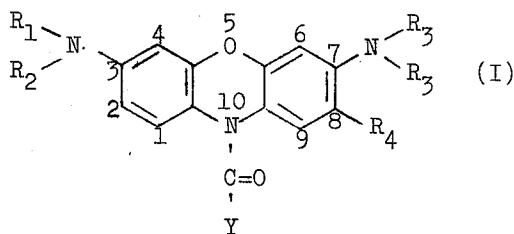
### EXAMPLE 3

#### Preparation of Pressure-sensitive Copying Papers

A blend of 1 part of Color Former No. 6 and 1.5 parts of crystal violet lactone was processed in the same manner as described in Example 1 to form a liquid containing microcapsules which was then applied to a paper and dried in the same manner as described in Example 1 to obtain a colorless upper sheet. When the upper sheet was superposed on a lower sheet coated with an acidic organic polymer, active clay substance or mixture thereof as a solid acid and localized pressure was applied to the assembly of sheets by, e.g., handwriting, a blue color was formed immediately in the pressed areas of the lower sheet. No substantial discoloration of the blue color was observed when the lower sheet was exposed directly to sun light for a long period of time. While the invention has been described in detail and in terms of specific embodiments thereof, it will be apparent to those skilled in the art that various modifications and variations can be made therein without departing from the spirit and the scope of the invention.

What is claimed is:

1. A pressure-sensitive copying paper comprising on a support a layer of an encapsulated color former and on the same or a different support a layer of an electron accepting solid, said color former being a phenoxazine compound represented by the formula



wherein R<sub>1</sub> and R<sub>3</sub> each represents a lower alkyl group; R<sub>2</sub> is a member selected from the group consisting of a lower alkyl group and a phenyl group; R<sub>4</sub> is a member selected from the group consisting of a hydrogen atom

and a methyl group; and Y is a member selected from the group consisting of an alkyl group, an alkenyl group, a styryl group, a benzyl group, a phenoxymethylene group, a phenyl group, a naphthyl group, a furyl group, a thienyl group, a pyridyl group and a quinolyl group, wherein the styryl, benzyl, phenoxymethylene, phenyl, and naphthyl groups may be substituted in the nucleus thereof by a substituent selected from the group consisting of a lower alkyl group, a lower alkoxy group, a di-lower-alkylamino group, a hydroxy group, a chlorine atom and a nitro group and wherein the quinolyl group may be substituted in the 2-position thereof by a substituent selected from the group consisting of a lower alkyl group and a phenyl group.

2. The pressure-sensitive copying paper according to claim 1, wherein said color former is a compound selected from the group consisting of 3,7-bis(diethylamino)-10-acetylphenoxazine, 3,7-bis(diethylamino)-10-crotonoylphenoxazine, 3,7-bis(diethylamino)-10-cinnamoylphenoxazine, 3,7-bis(diethylamino)-10-(p-nitrophenylacetyl)-phenoxazine, 3,7-bis(diethylamino)-10-(p-chlorophenoxyacetyl)-phenoxazine, 3,7-bis(diethylamino)-10-benzoylphenoxazine, 3,7-bis(diethylamino)-10-(p-methylbenzoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(p-methoxybenzoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(p-dimethylaminobenzoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(o-hydroxybenzoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(p-nitrobenzoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(2-furoyl)-phenoxazine, 3,7-bis(diethylamino)-10-(2-thienoyl)-phenoxazine, 3,7-bis(diethylamino)-10-nicotinoylphenoxazine, 3,7-bis(diethylamino)-10-isonicotinoylphenoxazine, 3,7-bis(diethylamino)-10-(2-phenylcinchonoyl)-phenoxazine, 3-diethylamino-7-dimethylamino-8-methyl-10-(p-methylbenzoyl)-phenoxazine, 3-diethylamino-7-dimethylamino-8-methyl-10-(1-naphthoyl)-phenoxazine, 3-diethylamino-7-dimethylamino-8-methyl-10-(3-hydroxy-2-naphthoyl)-phenoxazine, 3-diethylamino-7-di-n-butylamino-10-(p-methoxybenzoyl)-phenoxazine, 3-(N-methyl-N-phenylamino)-7-diethylamino-10-(p-methoxybenzoyl)-phenoxazine and a mixture thereof.

\* \* \* \* \*

UNITED STATES PATENT OFFICE  
CERTIFICATE OF CORRECTION

PATENT NO. : 3,873,340

DATED : March 25, 1975

INVENTOR(S) : Yoshihide Miyazawa et al

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

IN THE HEADING:

Insert --Foreign Application Priority Data

August 2, 1971 Japanese. .... . 57507/71

Signed and sealed this 20th day of May 1975.

(SEAL)

Attest:

RUTH C. MASON  
Attesting Officer

C. MARSHALL DANN  
Commissioner of Patents  
and Trademarks