# United States Patent [19]

**Jenkins**

[11] **3,803,074**

[45] **Apr. 9, 1974**


[54] **COLOUR REACTING COMPONENTS**


[75] Inventor: **Frank Llewellyn Jenkins,** Chesham, England

[73] Assignee: **Wiggins Teape Research & Development Ltd.,** London, England



[22] Filed: **Feb. 1, 1971**

[21] Appl. No.: **111,712**

[52] **U.S. Cl.** ..... **260/29.7 M,** 8/2.5, 117/36.2, 117/36.8, 117/36.9, 117/100 S, 117/152, 423/600

[51] **Int. Cl.** ..... **C08d 7/18,** B41m 5/22

[58] **Field of Search** ..... 117/36.2, 36.8, 36.9, 100 S, 117/152; 260/29.7 M, 29.7 S; 8/2.5; 106/292; 423/600; 252/463



[56] **References Cited**

UNITED STATES PATENTS

| | | | |
|---|---|---|---|
| 3,627,787 | 12/1971 | Han-Lin | 260/335 |
| 3,634,121 | 1/1972 | Brockett | 117/36.2 |
| 3,020,171 | 2/1962 | Bakan et al. | 117/36.2 |
| 3,226,252 | 12/1965 | Hemstock | 117/36.2 X |

OTHER PUBLICATIONS

Mathews, Application of Dyestuffs, pp. 166–169. (John Wiley & Sons, 1920)



*Primary Examiner*—Allan Lieberman
*Assistant Examiner*—T. DeBenedictis, Sr.
*Attorney, Agent, or Firm*—John P. Snyder



[57] **ABSTRACT**

A colour reactant component for coating a receptor sheet of the kind used in copying paper systems is produced by incorporating in an aqueous slurry of an inorganic mineral at least one soluble salt of the metals cobalt, cerium, copper, iron, manganese, lead, indium, lanthanum, barium, antimony, beryllium, strontium, nickel, zinc, calcium cadmium, magnesium, chromium, and tin, and at least one soluble aluminate salt, so as to precipitate at least one insoluble aluminate compound in the slurry, and thereafter incorporating a binding agent selected to bond the colour reactant component to the receptor sheet.


**17 Claims, No Drawings**

# COLOUR REACTING COMPONENTS

## BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to a method of producing colour reactant components for use with copying papers of the kind in which copying is effected by reacting together a colour former component and a colour reactant component, supported by a sheet of copying paper, herein referred to as a receptor sheet, to make a mark thereon, and particularly to the receptor sheets and the colour reactant components supported thereon.

2. Description of the Prior Art

In known copying papers the colour former component may be in solution or suspension and premature development of the colour former is prevented by encapsulation or by incorporating the colour former solution-suspension as a stabilized emulsion. The colour reactants are acidic materials intimately bonded to the receptor sheet and are used to develop the colour formers from a colourless to a coloured form. Thus the colour former normally forms a coating of droplets of printing fluid on one face of a transfer sheet, incorporated or encapsulated in material which is rupturable by printing or writing pressures applied to the other side of the transfer sheet so as to release said printing fluid or colour former for transfer to the face of underlying receptor sheet bearing the colour reactant component to form a colour image thereon.

The colour reactants normally used are natural or synthetic inorganic minerals, usually acidic minerals such as silicate minerals, silica, alumina or silica/alumina and acidic resins. Many of the inorganic minerals used give good image intensity but the formed images often undergo a blue green colour transition during exposure to visible and ultra-violet light. Colour reactants of the acidic resin type can often overcome some of the aforesaid disadvantages but when used in their natural state or in the ground or powdered form the image intensity or initial image intensity, is not of acceptable commercial quality.

A main object of the present invention is to provide a method for producing an improved colour reactant component for use in copying paper systems, in which colour reactant component, the aforementioned disadvantages are minimised or eliminated.

The present invention is based on the discovery that the use of metal aluminate salt in the colour reactant component results in an improved receptor sheet which gives images having increased stability to visible and ultra-violet light over those obtained when receptor sheets are used having colour reactant components including the natural and synthetic inorganic minerals used hitherto.

## SUMMARY

According to the present invention there is provided a method of producing a colour reactant component for coating a receptor sheet of the kind used in copying papers systems in which copying is effected by reacting together a colour former component and a colour reactant component supported on a receptor sheet, which method includes the steps of incorporating in an aqueous slurry of an inorganic mineral at least one soluble salt of the metals cobalt, cerium, copper, iron, manganese, lead, indium, lanthanum, barium, antimony, beryllium, strontium, nickel, zinc, calcium, cadmium, magnesium, chromium and tin, and at least one soluble aluminate salt, so as to precipitate at least one insoluble aluminate compound in said slurry, and thereafter incorporating a binding agent selected to bond the colour reactant component to the receptor sheet.

In an embodiment of the invention the anion of the soluble metal salt is chosen from the halide, nitrate and sulphate ions.

Preferably sodium aluminate is used as a soluble aluminate salt.

An inorganic mineral which is selected to be inert towards the colour former, is preferred because of better rheology. A suitable inert inorganic mineral is kaolin. Calcium carbonate and mica, an illite clay, may also be used as inert inorganic minerals.

In a further embodiment of the invention, the inorganic mineral may be selected to act as a co-reactant, for example calcine alumina and high surface area silica.

The precipitation of the aluminate salt in the presence of the inorganic mineral increases the reactivity of the aluminate salts over ordinary ground co-reactants due either to the precipitate being adsorbed onto the surface of the inorganic mineral and taking up the surface area characteristic of the inorganic mineral, or the inorganic mineral acting as a dispersing media for the precipitate as it forms.

The inorganic mineral and precipitate may then be combined with the binder, for example a styrene butadiene latex, and coated directly onto a paper sheet to form the receptor sheet, or it may be filtered first to remove residual soluble salts from the composition before the addition of the binder and subsequent coating.



## DESCRIPTION OF THE PREFERRED EMBODIMENTS

The invention will now be further described with reference to the following examples which for purposes of comparison also describe colour reactant components which incorporate metal salts other than aluminates. In the Tables showing values of print intensity the lower the value the more intense the print.

EXAMPLE 1

A slurry having the following composition was prepared:

| | |
|---|---|
| Calcium chloride | 0.05 moles |
| Kaolin clay | 40 gms |
| Water | 60 mls |

and a solution of 0.05 moles sodium aluminate in 60 mls water was added to the slurry. The mixture was stirred until homogeneous and was found to have the following composition:

| | |
|---|---|
| Calcium aluminate | 0.05 moles |
| Kaolin clay | 40 gms |
| Water | 120 gms |

The pH of the mixture was then adjusted to 5 by the addition of a suitable quantity of hydrochloric acid. 15 mls of styrene butadiene latex containing 50 percent solids and whose pH had also been adjusted, to 5 by the

addition of hydrochloric acid was then added to the mixture.

The mixture so prepared was then coated onto a sheet of paper to form a receptor sheet. This composition was found to provide a receptor sheet surface which when used in conjunction with a transfer sheet supporting a leucodye in encapsulated form gave images having improved intensity and resistance to blue-green colour transition with respect to images obtained using known forms of receptor sheets.

EXAMPLE 2

Using the method described in Example 1 a homogeneous mixture was prepared having the following composition:

| | |
|---|---|
| Cobalt aluminate | 0.1 moles |
| Kaolin clay | 40 g |
| Water | 140 g. |

The pH was adjusted to 5.5. using hydrochloric acid and 10g of styrene butadiene latex whose pH had also been adjusted to 5.5 with hydrochloric acid was added to the mixture. By a similar method but using sodium silicate in place of sodium aluminate a colour reactant component was prepared having the following composition:

| | |
|---|---|
| Cobalt silicate | 0.1 moles |
| Kaolin clay | 40 g |
| Styrene butadiene latex | 10 g |
| Water | 140 g |

The mixtures so prepared were each coated onto a separate sheet of paper at a coating weight of 7 to 9 gms per square meter to form receptor sheets which were used in conjunction with conventional transfer sheets supporting a leucodye in encapsulated form to give images of which the print intensity was determined at a period of 60 seconds after formation.

The results were as follows:

| | 60 sec print intensity |
|---|---|
| Receptor sheet using cobalt aluminate | 73 |
| Receptor sheet using cobalt silicate | 90 |

EXAMPLE 3

The procedure described in Example 2 was repeated but with sodium aluminate and sodium silicate using a soluble cerous salt to give two mixtures of the following composition:

| | | |
|---|---|---|
| I | Cerous aluminate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |
| II | Cerous silicate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |

Both mixtures were prepared at pH 5.5

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec print intensity |
|---|---|
| Receptor sheet using cerous aluminate | 49 |
| Receptor sheet using cerous silicate | 58 |

EXAMPLE 4

The procedure described in Example 2 was carried out but using a soluble zinc salt in conjunction with the sodium aluminate and sodium silicate to give two mixtures of the following composition:

| | | |
|---|---|---|
| I | Zinc aluminate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |

This mixture was prepared at pH 5.0

| | | |
|---|---|---|
| II | Zinc silicate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |

This mixture was prepared at pH 7.0

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec print intensity |
|---|---|
| Receptor sheet using zinc aluminate | 40 |
| Receptor sheet using zinc silicate | 62 |

EXAMPLE 5

The procedure described in Example 4 was carried out but using a soluble magnesium salt in conjunction with sodium aluminate and sodium silicate to give two mixtures of the following composition:

| | | |
|---|---|---|
| I | Magnesium aluminate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |
| II | Magnesium silicate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |

Both mixtures were prepared at pH 3.0

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec print intensity |
|---|---|
| Receptor sheet using aluminate | 56 |
| Receptor sheet using silicate | 59 |

EXAMPLE 6

The procedure described in Example 2 was carried out but using a soluble calcium salt in conjunction with the sodium aluminate and sodium silicate to give two mixtures of the following composition:

| | | |
|---|---|---|
| I | Calcium aluminate | 0.2 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 160 g |

This mixture was prepared at pH 5.0

| | | |
|---|---|---|
| II | Calcium silicate | 0.1 moles |
| | Kaolin clay | 40 g |
| | Styrene butadiene latex | 10 g |
| | Water | 140 g |

This mixture was prepared at pH 7.0

When used in conjunction with a transfer sheet as described in Example 2, the following results were obtained:

| | 60 sec print intensity |
|---|---|
| Receptor sheet using calcium aluminate | 43 |
| Receptor sheet using calcium silicate | 55 |

The effect of the pH and concentration of metal salt in the colour reactant component composition made up for coating onto the sheet of paper to form the receptor sheet can be seen in the following Tables 1 and 2 in which the print intensity was determined at a period of 60 seconds after formation.

The following results were obtained for the metal salt calcium aluminate:

TABLE 1

| pH | moles precipitate/40 gms Kaolin clay Control 0 | 0.05 | 0.1 | 0.2 |
|---|---|---|---|---|
| 3 | 88 | 72 | 73 | 87 |
| 5 | 87 | 58 | 43 | 48 |
| 7 | 87 | 75 | 57 | 55 |
| 9 | 87 | 65 | 55 | 57 |

TABLE 2

| | Kaolin added before precipitation | Kaolin added after precipitation | |
|---|---|---|---|
| Precipitate | Print intensity After 48 hrs. | Print intensity After 60 secs. | After 48 hrs. |
| Zinc Silicate | 45 | 52 | 52 |
| Zinc Aluminate | 43 | 50 | 49 |

The reactivity of the precipitate is unaffected when left in slurry form for periods of at least 3 days at an ambient temperature of between 15° and 20°C.

The results obtained using the aluminate anion were compared with those obtained using other anions besides silicates in the form of precipitates of various insoluble zinc salts. The coating mixes and tests of the receptor sheets so produced were carried out using the procedure of Example 2 and the conditions of pH and precipitate concentration given in the following Table 3 were those which gave the optimum results for each precipitate.

TABLE 3

| Reactive Constituent of coating mix. | Precipitate concn. 40 g Kaolin | pH | 60 sec. Print Intensity |
|---|---|---|---|
| Zinc aluminate | 0.1 moles | 5 | 40 |
| Zinc silicate | 0.1 moles | 7 | 62 |
| Zinc phosphate | 0.5 moles | 7 | 75 |
| Zinc hydroxide | 0.2 moles | 9 | 66 |
| Zinc carbonate | 0.1 moles | 9 | 48 |
| Zinc borate | 0.5 moles | 7 | 61 |
| Zinc chromate | 0.1 moles | 3 | 52 |

EXAMPLE 7

Two mixes of the following composition were prepared:

| | | |
|---|---|---|
| I | Zinc aluminate precipitated in situ according to the invention | 0.1 moles |
| | Calcium carbonate | 40 gms. |
| | Styrene butadiene latex (50% solids) | 20 gms. |
| | Water | 140 gms. |
| | The mix was prepared at pH 5. | |
| II | As mix I but without the zinc aluminate. | |

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec. print intensity |
|---|---|
| Receptor sheet incorporating I | 48 |
| Receptor sheet incorporating II | 100 |

EXAMPLE 8

Example 7 was repeated but replacing calcium carbonate with mica in both mix I and mix II.

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | |
|---|---|
| Receptor sheet incorporating I | 63 |
| Receptor sheet incorporating II | 100 |

EXAMPLE 9

Two mixes of the following composition were prepared:

| | | |
|---|---|---|
| I | Zinc aluminate precipitated in situ according to the invention | 0.1 moles |
| | Calcined alumina | 40 gms. |
| | Styrene butadiene latex 50% solids | 20 gms. |
| | Water | 140 gms. |
| | The mix was prepared at pH 5. | |
| II | As mix I but replacing the 0.1 moles zinc aluminate with 16 gms kaolin. | |

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec. print intensity |
|---|---|
| Receptor sheet incorporating I | 62 |
| Receptor sheet incorporating II | mix uncoatable due to gritty nature of mineral. |

EXAMPLE 10

Example 9 was repeated but replacing the calcined alumina with high surface area silica in both mix I and mix II.

When used in conjunction with a transfer sheet as described in Example 2 the following results were obtained:

| | 60 sec. print intensity |
|---|---|
| Receptor sheet incorporating I | 42 |
| Receptor sheet incorporating II | 48 |

By using the receptor sheets which incorporate the reactant component of the invention it is possible to obtain initial print intensities which are superior to those obtained using conventional receptor sheets incorporating other metal salts.

I Claim:

**1.** A method of producing a colour reactant component for coating a receptor sheet of the kind used in copying paper systems in which copying is effected by reacting together a colour former component and a colour reactant component supported on a receptor sheet, which method includes the steps of incorporating in an aqueous slurry of an inorganic mineral at least one soluble salt of the metals cobalt, cerium, copper, iron, manganese, lead, indium, lanthanum, barium, antimony, beryllium, strontium, nickel, zinc, calcium, cadmium, magnesium, chromium and tin, and at least one soluble aluminate salt, so as to precipitate at least one insoluble aluminate compound in said slurry, and thereafter incorporating a binding agent selected to bond the colour reactant component to the receptor sheet.

**2.** A method according to claim **1**, wherein the slurry is prepared in the presence of a solution of said soluble metal salt.

**3.** A method according to claim **1**, wherein the anion of the soluble metal salt is chosen from the halide, nitrate and sulphate ions.

**4.** A method according to claim **1**, wherein sodium aluminate is used as a soluble aluminate salt.

**5.** A method according to claim **1**, wherein the binding agent is added to the slurry mix containing the precipitated aluminate compound.

**6.** A method according to claim **1**, wherein the slurry containing the inorganic mineral and aluminate precipitate is filtered to remove residual soluble salts, and is then mixed with the binding agent.

**7.** A method according to claim **1**, wherein the inorganic mineral is selected to be unreactive with the colour former.

**8.** A method according to claim **7**, wherein the inorganic mineral is a kaolin clay.

**9.** A method according to claim **1**, wherein the inorganic mineral is selected to be reactive with the colour former.

**10.** A method according to claim **1**, wherein the binding agent is a styrene butadiene latex.

**11.** A method according to claim **2**, wherein the anion of the soluble metal salt is chosen from the halide, nitrate and sulphate ions, sodium aluminate is used as a soluble aluminate salt, and the binding agent is added to the slurry mix containing the precipitated aluminate compound after filtering of the slurry to remove residual soluble salts.

**12.** A method according to claim **11**, wherein the binding agent is a styrene butadiene latex.

**13.** The method of preparing a color reactor component to be applied to a receptor sheet which will exhibit improved image stability to visible and ultraviolet light when used with a color former component, said color reactor component consisting essentially of an aqueous slurry of an inorganic mineral, a metal aluminate precipitate in which the metal is selected from the group consisting of cobalt, cerium, copper, boron, manganese, lead, indium, lanthanum, barium, antimony, beryllium, strontium, nickel, zinc, calcium, cadmium, magnesium, chromium and tin, and binder; said method comprising:

forming said precipitate in said color reactor component by first forming an aqueous solution of a salt selected from the group consisting of halides, nitrates and sulphates of the metal corresponding to the precipitate, and then forming said precipitate by the addition of sodium aluminate to said salt solution.

**14.** The method according to claim **13** wherein said inorganic mineral is kaolin.

**15.** The method according to claim **13** wherein said inorganic mineral is calcium carbonate.

**16.** The method according to claim **13** wherein said inorganic mineral is calcined alumina.

**17.** The method according to claim **13** wherein said inorganic mineral is silica.

* * * * *