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## LEUCAURAMINE DERIVATIVES

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1 Claim

## ABSTRACT OF THE DISCLOSURE

A Group IIA metal, optionally substituted ammonium, optionally substituted hydrazine, optionally substituted hydroxylamine, optionally substituted guanidine or heterocyclic base salt of N-(2-carboxyphenyl) leucauramine or N-(4-carboxyphenyl) leucauramine.

The salts may be used as color formers in hectographic copying processes.

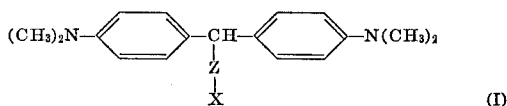
This invention relates to leucauramine derivatives of value in impact-printing systems using colorless carbon paper, spirit duplicating carbons and the like.

According to the invention, there are provided the Group IIA metal, optionally substituted ammonium, optionally substituted hydrazine, optionally substituted hydroxylamine, optionally substituted guanidine or heterocyclic base salts of N-(2- or 4-carboxyphenyl) leucauramine.

Particularly useful Group IIA metal salts are calcium salts. The substituted ammonium salts may be primary, secondary or tertiary amine salts or quaternary ammonium salts.

In general the preferred salts are the optionally substituted hydrazine, optionally substituted hydroxylamine, optionally substituted guanidine and heterocyclic base salts and particularly ammonium or substituted ammonium salts, especially those derived from  $RNH_2$ ,  $R_2NH$  or  $R_3N$  wherein R is alkyl, especially lower alkyl.

The salts of the invention may be prepared by reacting N-(2- or 4-carboxyphenyl)leucauramine with a Group IIA metal hydroxide, alkoxide, carbonate or bicarbonate or an appropriate nitrogenous base. The carboxy compounds, which are known compounds, may themselves be prepared by reacting anthranilic acid or p-aminobenzoic acid with a compound of the formula:—



wherein Z represents sulphur or, preferably, oxygen and X represents alkyl or, preferably, hydrogen. Examples of compounds of Formula I include Michler's Hydrol. Alternatively, the salts may be prepared directly by reacting a compound of Formula I with an appropriate salt of anthranilic acid or p-aminobenzoic acid.

The salts of the invention singly or as mixtures are particularly suitable for use in the production of carbon papers of the clean-to-handle type.

Carbon papers of the clean-to-handle type for use in the so-called hectographic or spirit-reproducing copying process consisting of tissue or other suitable film or sheet material on which is a coating containing a colorless derivative of a basic dyestuff such as Crystal Violet Lactone have already been proposed in British Patent 785,768. In the copying process the carbon paper is placed with its coated surface against one surface of a master paper

which is then typed, written or marked on causing transfer of the coating as a substantially colorless reverse image to the first-mentioned surface of the master paper at the points where carbon and master papers have been pressed together. The master paper is then brought into contact with a succession of sheets of paper moistened with a suitable spirit-reproducing fluid such as ethanol. The fluid dissolves a part of the basic dyestuff derivative and transfers it to each paper sheet where it combines with an activating substance such as an acid to give a visible color which will reproduce the original typing or writing on the master paper.

Color-formers hitherto proposed in this process have the disadvantage that they tend to give sticky coatings which are not easy to apply satisfactorily to the tissue or other film or sheet material and may give rise to unclear copies or copies having poor fastness to light. These disadvantages are decreased by the use of the salts of the present invention. The said salts may be included in coating compositions which may be applied by conventional means to a support material to produce the said clean-to-handle carbon papers.

The invention is illustrated but not limited by the following Examples in which all parts are by weight.

## EXAMPLE 1

To 20 parts of N-(2-carboxyphenyl) leucauramine in 100 parts of toluene are added, at room temperature, 6 parts of triethylamine and the resultant solution stirred for 10 minutes. A precipitate is formed which is isolated by filtration to give 17 parts of the triethylamine salt of N-(2-carboxyphenyl)leucauramine.

## EXAMPLE 2

To 50 parts of concentrated ammonia (S.G.O. 88) are added 15 parts of N-(4-carboxyphenyl) leucauramine with stirring. The off white precipitate which results is isolated by filtration to give 12 parts of the ammonium salt of N-(4-carboxyphenyl)leucauramine.

## EXAMPLE 3

To 1000 parts of saturated calcium hydroxide solution are added 3.89 parts of N-(2-carboxyphenyl)leucauramine with stirring and the reaction mixture stirred for 4 hours. The bluish-white solid which results is isolated by filtration and air dried to give 4 parts of the calcium salt of N-(2-carboxyphenyl) leucauramine.

## EXAMPLE 4

To a solution of 3 parts of di-o-tolylguanidine in 100 parts of water are added 3.89 parts of N-(2-carboxyphenyl)leucauramine with stirring to give a white precipitate. The solid is isolated by filtration and air dried to give 5.2 parts of the di-o-tolylguanidine salt of N-(2-carboxyphenyl) leucauramine.

## EXAMPLE 5

To 43 parts of Michler's Hydrol (62.3% paste) in 200 parts of toluene are added 21.6 parts of trimethylammonium anthranilate and the reaction mixture heated to reflux with stirring for 1 hour. On cooling a pale blue solid is deposited and is collected by filtration and air dried to give 30 parts of the trimethyl ammonium salt of N-(2-carboxyphenyl) leucauramine.

Each of the salts prepared as described in Examples 1-5 may be applied in conventional manner in the form of a coating composition to a support material to form a clean-to-handle carbon paper suitable for use in a spirit-duplicating process.

The following Table gives further Examples of salts which can be prepared in similar manner from Michler's Hydrol and which may be used in the preparation of clean-to-handle carbon papers.

| Ex. No. | Amine                | Salt                              | Method of preparation | Solvent  |
|---------|----------------------|-----------------------------------|-----------------------|----------|
| 6.      | p-Aminobenzoic acid. | N-methyl piperidinium             | Via free acid.        | Benzene. |
| 7.      | do.                  | Tetramethylhydrazinium            | do.                   | Toluene. |
| 8.      | do.                  | Morpholinium                      | do.                   | Do.      |
| 9.      | do.                  | Ethyltrimethylammonium            | do.                   | Do.      |
| 10.     | do.                  | Ethylammonium                     | do.                   | Do.      |
| 11.     | do.                  | Triethylammonium                  | Direct (see Ex. 5)    | Ethanol. |
| 12.     | do.                  | N-methyl-O-methylhydroxylammonium | Via free acid.        | Toluene. |
| 13.     | do.                  | Magnesium                         | do.                   | Water.   |
| 14.     | do.                  | Calcium                           | do.                   | Do.      |
| 15.     | do.                  | Diethylammonium                   | do.                   | Toluene. |
| 16.     | do.                  | t-Butylammonium                   | do.                   | Do.      |
| 17.     | do.                  | Tetramethylammonium               | do.                   | Do.      |
| 18.     | Anthranilic acid.    | Diethylammonium                   | do.                   | Do.      |
| 19.     | do.                  | Strontium                         | do.                   | Water.   |
| 20.     | do.                  | N,N'-diethylpiperazinium          | do.                   | Toluene. |
| 21.     | do.                  | Dimethylphenylhydrazinium         | do.                   | Do.      |
| 22.     | do.                  | Barium                            | do.                   | Water.   |

#### We Claim:

1. A primary, secondary or tertiary lower alkyl amine salt of N-(2-carboxyphenyl) leucauramine or N-(4-carboxyphenyl) leucauramine.

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