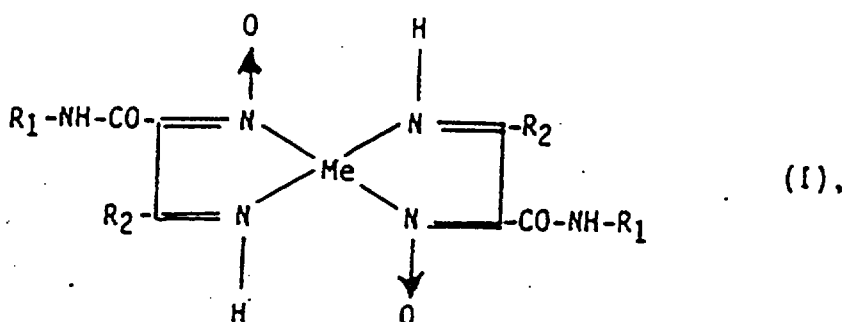


1:2 Metal complexes useful as pigments

The invention relates to novel 1:2 metal complex compounds that are useful as pigments.

According to the invention, there is provided a compound of
5 formula I



in which each R_1 independently, is a single ring or fused ring aromatic or heteroaromatic group, free of water-solubilising groups;

each R_2 independently is phenyl or methyl; and
 Me is Ni, Cu, Co or Zn.

10 Preferably R_1 is R_{1a} where R_{1a} is phenyl, unsubstituted or substituted by 1 to 3 substituents selected from chloro, bromo, methyl, methoxy and ethoxy or benzimidazole unsubstituted or monosubstituted by chloro, methyl or methoxy.

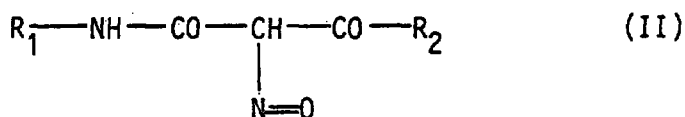
Preferably R₂ is methyl.

Preferably both groups R₁ are the same

Preferably both groups R₂ are the same.

Preferably Me is Ni.

- 5 Further, according to the invention, there is provided a process for preparing a compound of formula I defined above comprising aminating and metallising 2 moles of a compound of formula II



- 10 with 1 mole of a water-soluble Ni, Cu, Co or Zn salt in ammonia-alkaline medium at a temperature from 50 to 100°C inclusive.

The compounds of formula II are known or can be prepared by known methods from known compounds.

- 15 Amination and metallisation of a compound of formula II is generally carried out in water but may also be carried out in water-soluble organic solvents such as lower alcohols, glycols and glycol ethers and in mixture of these solutions in water.

- 20 The compounds of formula I can, according to known methods, be heat treated with high boiling point organic liquids, such as DMF (dimethylformamide), in which the compounds are not soluble, so as to purify the compounds so as to improve their pigment properties.

- 25 The compounds of formula I are suitable for the mass pigmenting of synthetic plastics and resins, free from or containing solvents, e.g. viscose, cellulose acetate, polyethylene, polystyrene, polyvinyl chloride and synthetic leathers and rubbers, of surface coatings, such as paints, whether oil or water based, lacquers and inks. They may also be used in pigment printing, textile

coating and for pigmenting paper in the mass. They may be employed for such uses in conventional manner and in conventional amounts. They are particularly useful in the pigmentation of paints and lacquers.

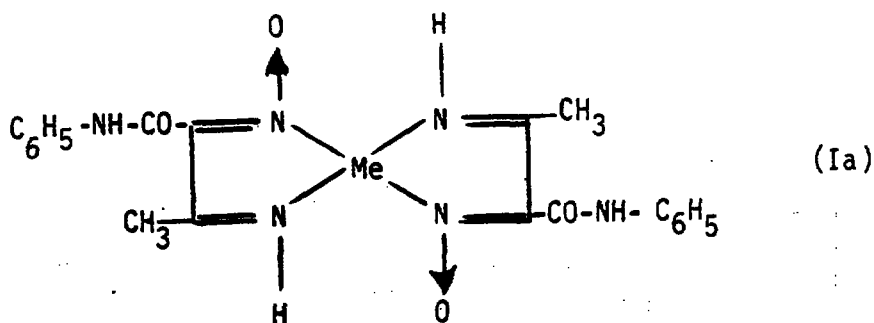
- 5 The resulting dyeings incorporating compounds of formula I have good heat, light and weather fastness. They also have a good stability to chemicals and have good (low) migration properties e.g. resistance to surface blooming fastness and to solvents for migration when overcoatings are applied.

- 10 The compounds of formula I have a good dye strength and have good application properties such as good anti-flocculation and anti-crystallisation properties and good dispersibility.

The invention will now be illustrated by the following Examples in which all parts and percentages are by weight and all temperatures are in °C.

EXAMPLE 1

70.8 Parts of acetoacetanilide are added, with stirring to a mixture of 300 parts of water and 60 parts of glacial acetic acid. The mixture is cooled to 5°, reacted with 28 parts of a 4N aqueous solution of sodium nitrite and stirred for 12 hours at 5 to 10°. The resulting nitroso compound (of formula II) is filtered off, washed with cold water, added to 200 parts of 25 % ammonia solution and reacted with a solution of 51.2 parts of nickel acetate tetrahydrate in 300 parts of water. The mixture is stirred at 80° for 12 hours and the product, which is the compound of formula Ia

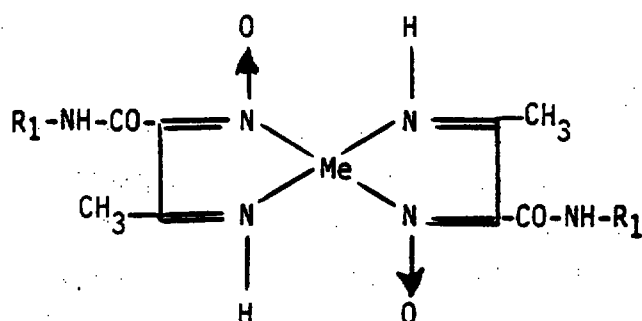


is hot filtered, washed with water and dried.

The resulting pigment is then refluxed for about 30 minutes in DMF (dimethylformamide) to purify the product and the product is then filtered, cold washed with methanol and dried. The product produces a yellow nuance in the substrate to be pigmented.

EXAMPLES 2 TO 12

Compounds of the formula



in which both groups R_1 are the same and are as defined in the Table below, can be prepared by a method analogous to that of Example 1 from appropriate reactants.

TABLE

Ex. No.	R_1	Colour of resulting pigmentation
2	2,5-Dimethoxyphenyl	yellow
3	2,4-Dimethoxyphenyl	orange
4	2-Methoxyphenyl	yellow-orange
5	2-Chlorophenyl	orange
6	2-Methylphenyl	do.
7	4-Methylphenyl	do.
8	2,4-Dimethylphenyl	yellow-orange
9	2,4-Dichlorophenyl	yellow
10	4-Methoxyphenyl	orange
11	Benzimidazol yl-5	bordeaux-red
12	4-chlorophenyl	orange

EXAMPLES 13, 14 and 15

Example 12 is repeated, using instead of nickel acetate, an appropriate amount of copper acetate (Example 13), cobalt acetate (Example 14) or zinc chloride (Example 15). Pigmentations in
5 cocoaldehydemelamine resins made with the pigment of Examples 13 and 14 give a brown nuance and with that of Example 15 a light yellow nuance.

EXAMPLE 16

Example 1 is repeated using 109.2 parts of benzoylacetanilide
10 instead of the 70 parts of acetoacetanilide.

The resulting pigment colours cocoaldehyde-melamine resin lacquers yellowish brown.

APPLICATION EXAMPLE A

15 4 Parts of the pigment of Example 1 are milled with 96 parts of a mixture of

50 parts of a 60 % solution of coco-aldehyde-melamine resin (32 % fat content) in xylene;

20 30 parts of a 50 % melamine resin solution in butanol;
10 parts of xylene and

10 parts of ethyleneglycol monoethylether
in a ball mill for 24 hours.

The resulting dispersion is sprayed onto an aluminium piece, left to air dry for 30 minutes and is then stoved for 30 minutes at 120°. A
25 brilliant yellow film, with good light and weather fastness properties results.

APPLICATION EXAMPLE B

A PVC sheet containing 0.1 % pigment (average ratio coloured pigment to white pigment 1:5) can be prepared as follows:

30 16.5 parts of a softener mixture of equal parts dioctylphthalate and dibutylphthalate are mixed with 0.05 parts of a pigment of Example 1 and 0.25 parts of titanium dioxide. 33.5 Parts of polyvinylchloride are then added.

The mixture is mixed for 10 minutes between 2 rollers in a rolling machine, regularly cuttin and rerolling the resulting sheet. One of the rollers is held at a temperature of 40° and the other at a temperature of 140°.

- 5 The mixture is then as a sheet and is pressed between two polished metal plates at 160° for 5 minutes.

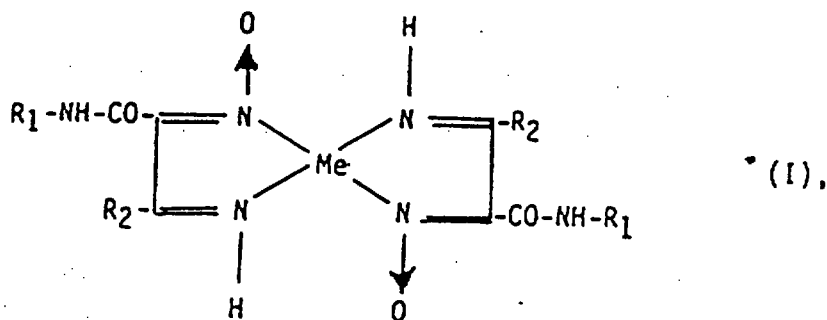
A yellow PVC sheet having a high brilliance, and very good migration good and light fastness results.

- 10 In application Examples A and B, an appropriate amount of any one of the pigments of Examples 2 to 16, may be used in place of the pigment of Example 1.



CLAIMS: -

1. A compound of formula I



in which each R_1 independently, is a single ring or fused ring aromatic or heteroaromatic group, free of water-solubilising groups;

5

each R_2 independently is phenyl or methyl; and

Me is Ni, Cu, Co or Zn.

2. A compound according to Claim 1, in which R_1 is R_{1a} where R_{1a} is phenyl, unsubstituted or substituted by 1 to 3 substituents selected from chloro, bromo, methyl, methoxy and ethoxy or benzimidazole, unsubstituted or monosubstituted by chloro, methyl or methoxy.

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3. A compound according to Claim 1 or Claim 2, in which R_2 is methyl.

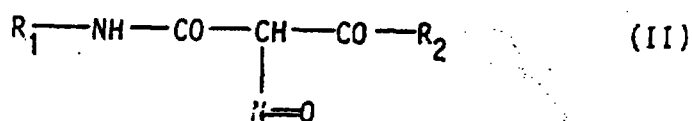
4. A compound according to any one of the preceding claims in which Me is Ni.

15

5. A compound according to Claim 1, substantially as herein described with reference to any one of Examples 1 to 16.

6. A process for preparing a compound of formula I defined in Claim 1 comprising aminating and metallising 2 moles of a compound of formula II

20



with 1 mole of a water-soluble Ni, Cu, Co or Zn salt in ammonia-alkaline medium at a temperature from 50 to 100°C inclusive

7. A process according to Claim 6, substantially as herein described with reference to any one of Examples 1 to 16.

5 8. A substrate to which a compound according to any one of Claims 1 to 5 has been applied.

9. A process for pigmenting a substrate comprising applying to that substrate a compound according to any one of Claims 1 to 5.

10. A substrate when pigmented according to Example A or B.
