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FLUORESCENT COMPOSITION CONTAINING A COUMARONE-INDENE RESIN

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ABSTRACT OF THE DISCLOSURE

This invention relates to compositions of matter useful for marking and identification purposes comprising a solvent, a fluorescent material and a coumarone-indene resin and a method of marking and identifying objects therewith.

BACKGROUND OF THE INVENTION

One of the basic problems which has plagued military personnel over the years has to do with the identification of objects. This problem relates not only to personnel but also to equipment such as trucks, boats, etc. That is to say, in recent years it has become more and more difficult to identify insurgents by their clothing, appearance, etc. and therefore a system of identification whereby it is possible to determine whether a subject in question has come through a specific military zone has become necessary. Many known systems have been tried, however, they fail because of various deficiencies not the least of which is that they require extensive and cumbersome equipment in order that they may be detected. Such equipment is obviously impossible to transport and utilize, especially in the field of combat. Analogous systems are taught in U.S. Pat. Nos. 3,162,642 and 3,066,105.

SUMMARY

I have now discovered a class of compositions which may be applied to various objects via a multiplicity of methods for purposes of identification and detection of insurgents. My compositions are featured by their increased quantum efficiency. The intensity of emission of my compositions is much greater than that of known compositions when examined under a long wave ultraviolet light, and as intrusion control markers, they may be readily applied to foliage or the ground and detected at a later time on persons who have traversed a given area illegally.

Prior art systems such as non-wettable powders and oils fail to meet accepted military requirements, as do conventional dyes such as those used to trace the flow of rivers or mark the location of survivors of maritime disasters. The nonwettable powders adhere poorly to the surface of the objects and therefore are easily removed, while the oils tend to penetrate too deeply into porous objects and therefore become undetectable. The dyes are too easily visible by the human eye and therefore objects marked therewith can be easily avoided.

DESCRIPTION OF THE INVENTION INCLUDING PREFERRED EMBODIMENTS

As mentioned above, my novel compositions comprise a non-fluorescent solvent, a fluorescent material and a coumarone-indene resin.

Examples of suitable solvents useful in my novel compositions include the aromatic hydrocarbons such as xylene, toluene, benzene, etc. normally liquid aliphatic hydrocarbons such as pentane, hexane, heptane, etc. ethyl acetate, ethyl chloride, butyl formate, dibutyl ether, methylethyl sulfide, methylbenzyl ether, water, substantially aromatic petroleum fractions, e.g. cycle oil, fuel oil, etc. The amount of solvent used in my compositions

is not critical except that enough should be used so as to dissolve the other components thereof.

The fluorescent material used in my compositions can consist of any material or combination of materials which can be activated by ultraviolet light and which emit in the visible (are colored in the visible region). The fluorescent material can be utilized in amounts ranging from about 0.1% to about 5.0%, by weight, based on the total weight of the final composition. The fluorescent material is preferably soluble in the solvent and the resinous material but may also be utilized as a suspension therein. Examples include the benzothiazoles (U.S. 3,049,509; 2,985,661) e.g.

2-(2-hydroxy-5-chlorophenyl)-benzothiazole;
2-(2-hydroxy-3-methoxyphenyl)-benzothiazole;
2-phenyl-5-methyl-6-methoxy benzothiazole;
2-(2-aminophenyl)-benzothiazole;
2,2′-bis-benzothiazole;
2-benzoimido-6-methoxy-3-methylbenzothiazole;
2-(p-acetamidophenyl)-6-methylbenzothiazole,

etc.; the quinazolones (U.S. 3,169,129; 3,269,955; 3,526,627) e.g.

2-(2-hydroxy-3,5-dichlorophenyl)-4(3)-quinazolone;
2-(4-methoxy-phenyl)-4(3)-quinazolone;
6-methoxy-2-phenyl-4(3)-quinazolone;
6-butyl-2-(4-methoxyphenyl)-4(3)-quinazolone;
2-(3-hydroxy-2-naphthyl)-quinazolone;
1,2-dihydro-2-(*o*-methoxyphenyl)-quinazolone;
2-(2-hydroxyphenyl)-quinazolone ethyl carbonate;
2-styryl quinazolone; 2-(2-hydroxy-3-nitrophenyl)-4(3)-quinazolone;
2-[2-hydroxy-5-(azophenyl)phenyl]-4(3)-quinazolone,

etc.; 4-methyl-7-dimethylamino coumarin (U.S. 2,844,539); the oxazoles, e.g. 2-(1-naphthyl)-5-phenyl oxazole (U.S. 2,985,661); the benzimidazoles (U.S. 2,985,661) e.g.

2-(2-hydroxyphenyl)-benzimidazole; 2 - phenylbenzimidazole; 2 - (2-aminophenyl)-benzimidazole; etc.; the hydrazines e.g. disalicyloylhydrazine; 1-salicyloyl 2-(4-methoxy salicyloyl)hydrazine; 1,2-*bis* - (3,5-dichloro salicyloyl)hydrazine,

etc., also such classes of compounds represented by such compounds as 1,8-dihydroxyanthraquinone; 4,5-diphenylimidazolone; 2 - (*o*-hydroxyphenyl)benzoxazole (U.S. 2,985,661); N,N″ - *bis*-(salicylideneamino)oxamidene (U.S. 3,149,092); p-methoxy-phenyl hydrazone; 1-cyanoformimidic acid; alpha - methyl-*o*-hydroxy - benzylidenehydrazide (U.S. 3,149,092); 6-hydroxynicotinic acid; salicylaldehyde semi-carbazone; 2-aminobiphenyl; 2-phenylbenzoxazole (U.S. 2,985,661); bis(8 - hydroxyquinolino) Zn II, and the like.

The coumarone-indene should be present in amounts ranging from 50% to about 90%, preferably about 55% to about 85%, by weight, based on the total weight of the final composition. Any coumarone-indene resin (80:20; 20:80) system may be utilized, classes of such resins and methods for their production being well known to those skilled in the art.

The compositions of the instant inventions are prepared by merely mixing the ingredients together in the appropriate amounts. No critical mixing features are necessary. If a dispersion is used, the ingredients must merely be suspended before application.

As mentioned above, my novel compositions may be used to mark many objects including humans, equipment such as trucks, boats, guns, ammunition, etc. clothing, trees, shrubs and bushes etc. They may also be used as a detecting mechanism whereby one can readily determine

an area previously covered or patrolled, etc. such as daily aerial spraying, as mentioned above.

My novel compositions may be detected after application by merely contacting the object in question with ultraviolet light and examining the contacted area in the dark with the human eye.

The following examples are set forth for purposes of illustration only and are not to be construted as limitations on the instant invention except as set forth in the appended claims. All parts and percentages are by weight unless otherwise specified.

EXAMPLE 1

To a suitable vessel are added 24 parts of xylene. 75.0 parts of a commercially available coumarone-indene resin are then added to the vessel and stirred until dissolved. To the resultant mixture is then added 1.0 part of 2-(2-hydroxyphenyl)benzothiazole. The ingredients are stirred until a clear solution is obtained.

EXAMPLE 2

Following the procedure of Example 1 except that an equivalent amount of 2-(2-hydroxy-3,5-dichlorophenyl)-4-(3H)-quinazolone is substituted for the benzothiazole, a similar composition is produced.

EXAMPLE 3

Again following the procedure of Example 1 except that an equivalent amount of 2-(2-hydroxyphenyl)-6-chloro-4-(3H)-quinazolone is used in place of the benzothiazole, a similar composition is produced.

EXAMPLE 4

(Comparative)

Again following the procedure of Example 1 except that 59 parts of a commercially available mixture of alkyd resins and 40 parts of xylene is utilized in place of the coumarone-indene resin.

EXAMPLE 5

(Comparative)

The procedure of Example 1 is again followed except that an equivalent amount of a commercially available phenol-formaldehyde resin is substituted for the coumarone-indene resin thereof.

The compositions of Examples 1–5 were each then tested to determine the quantum efficiency thereof. A Bechman® DK–2A instrument was used to measure the reflectance and transmission spectra with appropriate "cut-off" filters mounted in front of the detector to remove fluorescence.

The instrumentation and calculations used to determine the luminescence efficiency are described in Applied Spectroscopy, vol. 21, No. 4, p. 250. The reference material used was the National Bureau of Standards phosphor No. 28. Quinine sulfate of 1×10^{-3} molar in 0.1 N H_2SO_4 was used exclusively for calibration checks. The results obtained on excitation with light of 2570 A. wavelength are set forth in Table I, below.

TABLE I

Composition of example number:	Luminescence efficiency
1	0.07
2	0.06
3	0.06
4	0.04
5	0.04

It can therefore be seen that my novel compositions have a luminescence efficiency of 150–175% greater than compositions utilizing an alkyd or phenol-formaldehyde resin.

The compositions of Examples 1–3 were then sprayed on foliage using a back pack sprayer powered by a small gasoline engine. The marked area was then traversed by a person wearing a typical uniform made from black cotton fabric. Subsequent examination of the clothing in the dark and detection of the markings was made by shining a longwave ultraviolet light on the material. The markings were still observable 2 to 3 weeks later.

EXAMPLES 6–13

Substitution of the following fluorescent materials for that of Example 1 resulted in compositions which function as that of said example, both in quantum efficiency and stability.

6—2-phenyl-5-methyl-6-methoxybenzothiazole;
7—2-(2-hydroxyphenyl)-quinazolone ethyl carbonate;
8—4-methyl-7-dimethylamino courmarin;
9—2-(1-naphthyl)-5-phenyl oxazole;
10—2-phenylbenzimidazole;
11—1,2-bis(3,5-dichlorosalicyloyl)hydrazine;
12—1,8-dihydroxyanthraquinone;
13—2-(o-hydroxyphenyl)benzoxazole.

What is claimed is:

1. A composition of matter comprising (A) a non-fluorescent solvent for (B) and (C), (B) a fluorescent material which is activated by ultraviolet light and which emits in the visible region and (C) a coumarone-indene resin.

2. A composition according to claim **1** wherein (A) is xylene.

3. A composition according to claim **1** wherein (B) is 2-(2-hydroxyphenyl)benzothiazole.

4. The composition of claim **1** wherein (B) is 2-(2-hydroxyphenyl)-6-chloro-4-(3H)-quinazolone.

5. The composition of claim **1** wherein (B) is 2-(2-hydroxy-3,5-dichlorophenyl)-4-(3H)-quinazolone.

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