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(54) Title: AN IMPROVED ADDITIVE FOR IMPARTING BACTERICIDAL AND ANTIMICROBIAL PROPERTIES TO A MA-
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(57) Abstract: An additive for imparting bactericidal and antimicrobial properties to a material, such as a paint composition, the ad-
ditive comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof dissolved
in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt,
an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid. In
a preferred embodiment, the carrier is a triglyceride or a fatty acid ester having the general basic formula: R-O-CO-C_xH_y wherein R
is a branched or straight chain alkyl group having at least 3 carbon atoms; X is 13 to 17 and Y is 27 to 35.

WO 2004/071537 A2

Title: An Improved Additive for imparting bactericidal and antimicrobial properties to a material.

DESCRIPTION

The present invention relates to an improved additive for imparting bactericidal and antimicrobial properties to a material, particularly but not exclusively for use in surface coatings such as paints and sealants.

Triclosan or 2,4,4'-trichloro-2'-hydroxy diphenyl ether has been widely used since the 1960's against the spread of microbes. The compound has been incorporated into a range of consumer goods to impart bactericidal properties, including plastics for toys and kitchen utensils, cosmetics and toothpaste. The substance is a potent killer of microbes, such as bacteria and fungi and acts by blocking the active site of an enzyme called enoyl-acyl carrier-protein reductase ("ENR") which prevents the microbe from manufacturing fatty acids required for cell membrane construction and other vital functions. Humans do not possess this enzyme and therefore the compound is harmless to them thereby enabling its incorporation into consumer products as a safe bactericide.

Triclosan is a powder that is scarcely soluble in water. This limits the application of the compound to certain products. The powder has been provided as a dispersion in paint, but although satisfactory, this paint composition does suffer from a number of drawbacks. Firstly, the Triclosan powder can only be added at certain stages of the manufacturing process. Secondly, due to the particulate nature of the powder, the Triclosan may be distributed unevenly throughout the coating and may subsequently be removed from the surface by cleaning operations, causing paints based on a dispersion of Triclosan powder to experience loss of bactericidal

protection over time. Furthermore, attempts to dissolve the triclosan in solvents can give paints an undesirable granular appearance when applied to a surface. These problems have existed for a long period of time.

It is an object of the present invention to provide an additive for a material, particularly but not exclusively for a surface coating, such as a paint composition, that may be added to the surface coating during or after its manufacture to impart bactericidal and/or antimicrobial properties thereto.

It is a further object of the present invention to provide an improved surface coating composition, such as a paint composition, having bactericidal and/or antimicrobial properties that aims to overcome or at least alleviate the abovementioned drawbacks.

It is yet a further object of the present invention to provide a method for imparting bactericidal and/or antimicrobial properties to a material, such as a surface coating, that aims to overcome or at least alleviate the abovementioned drawbacks.

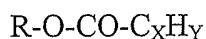
Accordingly, a first aspect of the present invention provides an additive for a material, the additive comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether or a derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.

Preferably, a long chain fatty acid and/or fatty acid ester and/or ester of glycerol is used, including triglycerides. Preferably, fatty acids or esters having chain lengths of 6 to 20 carbon atoms are used. Branched or straight chain, saturated, unsaturated, or polyunsaturated fatty acids may be used. Preferably, the

fatty acids are saturated. The fatty acids and/or fatty acid esters or salts may be natural or synthetic. Natural or synthetic oils or fats may be used as the source of the fatty acid and/or fatty acid esters or salts. Derivatives may include, for example, ethoxylated fatty acid esters.

The carrier for dissolving the 2,4,4'-trichloro-2'-hydroxy diphenyl ether may be one or more compounds selected from the group consisting of linseed oil, mineral oil, castor oil, soya oil, tall oil fatty acids, C₁₂-C₁₅ alkyl benzoate, MCT oil (medium chain triglycerides), 2-Ethyl Hexyl palmitate, 2-Di-Ethyl Hexyl adipate, triglyceride ester, isodecyl oleate, isopropyl myristate, isopropyl palmitate, isopropyl stearate and butyl myristate. The fatty acid may also be sodium, potassium and amine soaps of, for example, oleic, ricinoleic, coconut and myristic fatty acids. Preferably, the carrier is selected from the group consisting of at least one C₁-C₂₀ alkyl ester of a long chain alkyl or aryl carboxylic acid, including fatty acids, dicarboxylic acids and benzoic acids or a glycerol ester.

More preferably still, the carrier for the 2,4,4'-trichloro-2'-hydroxy diphenyl ether is a triglyceride, especially MCT oil, or a fatty acid ester having the general basic formula:



wherein R is a branched or straight chain alkyl group having at least 3 carbon atoms; X is 13 to 17 and Y is 27 to 35.

Preferably, R has 3 to 8 carbon atoms.

Preferred additives according to the present invention comprise 2,4,4'-trichloro-2'-hydroxy diphenyl ether and a fatty acid ester selected from the group

consisting of isopropyl myristate, 2-ethyl-hexyl palmitate, butyl myristate, isopropyl stearate and isopropyl palmitate.

The solution preferably contains a maximum of 60% by weight 2,4,4'-trichloro-2'-hydroxy diphenyl ether. Preferably, the solution contains a minimum of 30% by weight of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, more preferably 30% to 50%, especially 40% by weight. The solution may also contain other minor ingredients, such as dispersants/surface tension modifiers and additional microbiologically active substances (such as 2-n-octyl-4-isothiazolin-3-one). Preferably, the minor ingredients are in an amount 0 to 10% w/w, or in amount of 0-30% in the case of microbiologically active substances.

Preferably, the viscosity of the solution is 1mPa (0.01 Poise) to 2 Pas (20 Poise).

A second aspect of the present invention provides the use of an additive comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether or derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid for imparting bactericidal and/or antimicrobial properties to a material, such as a surface coating, particularly a paint formulation.

A third aspect of the present invention provides a method for imparting bactericidal and/or antimicrobial properties to a material comprising adding to the material 2,4,4'-trichloro-2'-hydroxy diphenyl ether or derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl

ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.

Preferably, the surface coating comprises a paint formulation. In a preferred embodiment of the present invention there is provided a paint composition comprising a binder and a pigment and further comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether or a derivative thereof dissolved in a carrier, the carrier comprising at least one from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.

Any appropriate binder, such as a resin or polymer, may be included in the paint composition as is standard in the art. Similarly, any pigment may be used, such as titanium dioxide. The paint composition may also contain solvents, thinners and diluents and may include conventional additives for paints, such as anticorrosive pigments, filler/extender pigments, dispersants, preservatives, thickening/structuring agents, antifoaming agents, antisetling agents and antiskinning agents.

The additive may be incorporated into other surface coatings such as sealants and mastics to impart bactericidal and antimicrobial properties to the coating. The solution may also be added to a cleaning solution.

The solution of the 2,4,4'-trichloro-2'-hydroxy diphenyl ether in a carrier as herein described may be added to a surface coating composition during or after manufacture of the coating.

Preferably, the solution of 2,4,4'-trichloro-2'-hydroxy diphenyl ether is present in the material to which it is added at less than 5% by volume, more preferably 0.1 to 2.5%, especially 0.5-1.5 %. It is to be appreciated that a material

may be provided in a concentrated form, which is diluted prior to application to a substrate.

The invention will now be further illustrated by means of the following Examples in which Example 1 describes the preparation of an additive according to the present invention and its incorporation into a surface coating, Examples 2 to 5 provide the compositions of different types of paint formulations provided with the additive of the present invention, Example 6 describes a mastic composition provided with the additive of the present invention, Example 7 describes a cleaning solution provided with the additive of the present invention and Example 8 investigates the bactericidal properties of paint formulations provided with the additive of the present invention, and with reference to the accompanying drawings in which:

Figure 1 is a plot of the number of viable bacteria against time (in days) for treated and untreated Paracem Semi-gloss paint; and

Figure 2 is a plot of the number of viable bacterial against time (in days) for treated and untreated Pegakote 2-pack epoxy paint.

Example 1.

40% by weight of Triclosan (2,4,4'-trichloro-2'-hydroxy diphenyl ether) was dissolved in 60% by weight of isopropyl myristate (synonyms include tetradecanoic acid 1-methylethyl ester, estergel, isopropyl tetradecanoate, myristic acid isopropyl ester). This solution was then added to a water-based paint composition in an amount 1-2% by volume. The Triclosan appears to be completely dispersed within the paint composition with no granular appearance.

Upon application of the paint to a surface, the Triclosan solution of the present invention appears to be adsorbed onto the surface of the paint resin. As the paint dries and water evaporates, the paint resin forms a film with the Triclosan forming an integral part of the film. This also suggests that, following scrubbing of the film, a diffusion process will replenish depleted levels of the antimicrobial substance within the surface coating. The solution form of the Triclosan also allows for its addition at any stage of the manufacture of the paint and even post-manufacture, allowing addition of the solution as an additive at or after the point of sale of the paint.

Example 2

The Triclosan solution according to the present invention was added to a waterborne 2-pack epoxy finish paint to provide the following composition:

<u>Component 1</u>	<u>% by Weight</u>
Polyamide adduct supplied 80% in water amine value (ISO 9702) 205mg KOH/g	17.00
Tertiary amine accelerator amine value (ISO 9702) 620mg KOH/g	1.00
Water	13.00
Titanium Dioxide	19.00
Talc	17.50
<u>Component 2</u>	
Bisphenol A/F liquid epoxy resin (emulsifiable) epoxy equivalent (ISO 3001) 177/g/Eq	24.50

Water	6.75
40% active triclosan solution	1.25
	100.00%

The Triclosan solution was added to Component 1 but could have been added to Component 2 to provide a paint composition having the required properties. The triclosan solution replaces water in the standard paint formulation.

Example 3.

The Triclosan solution according to the present invention was added to a waterborne matt emulsion paint to provide the following composition:

	<u>% by weight</u>
Water	28.00
Cellulose thickener	0.50
Defoamer	0.02
China Clay	9.00
Titanium Dioxide	22.50
Coalescing Agent	1.50
Water	8.76
50% Vinyl Acetate/VeoVa10 copolymer dispersion	27.50
Phthalate Plasticiser	0.75
In-can (wet-stage) preservative	0.20
Defoamer	0.02
40% active triclosan solution	1.25

100.00%

The triclosan solution replaces water in the basic paint formulation.

Example 3.

The Triclosan solution according to the present invention was added to a waterborne gloss emulsion paint to provide a composition having the following formula:

	% by weight
Propylene Glycol	7.00
Pigment Dispersant	1.00
Defoamer	0.35
In-can (wet stage) preservative	0.35
Titanium Dioxide	25.00
Water	2.05
50% Vinyl Acetate/VeoVa10 copolymer dispersion	63.00
40% active triclosan solution	1.25
	100.00%

The triclosan solution replaces water provided in the basic paint formulation.

Example 4

A triclosan solution of the present invention was added to a decorative quality gloss enamel paint for brush application to provide the following composition:

	<u>% by weight</u>
Linoleic rich long oil alkyd	20.00
70% solids, 64% fatty acid	
Titanium Dioxide	25.00
White Spirit	5.00
Linoleic rich long oil alkyd	30.00
70% solids, 64% fatty acid	
12% Cobalt Driers	0.20
18% Zirconium Driers	0.60
10% Calcium Driers	0.90
Anti Skinning Agent	0.10
White Spirit	16.95
40% active triclosan solution	1.25
	100.00%

The triclosan solution additive replaces white spirit provided in the basic enamel formulation.

Example 5.

A triclosan solution of the present invention was incorporated into a quick drying industrial enamel for spray application to provide the following composition:

	<u>% by weight</u>
Linoleic rich short oil alkyd	20.00
50% solids, 32% fatty acid	
Titanium Dioxide	20.00
Xylene	10.00
Linoleic rich short oil alkyd	32.00
50% solids, 32% fatty acid	
12% Cobalt Driers	0.20
18% Zirconium Driers	0.70
Anti Skinning Agent	0.10
Xylene	15.75
40% active triclosan solution	1.25
	100.00%

The triclosan solution of the present invention replaces an equivalent amount of Xylene in the basic enamel formulation.

Example 6.

A triclosan solution of the present invention was added a mastic formulation to provide the following composition:

	<u>% by weight</u>
Plasticised acrylic resins	40.00
Titanium Dioxide	2.00
Dolomite (extender pigment)	30.00

12

Additives (defoamers/thickeners/wetting agents)	1.00
Ammonia Solution	0.25
40% active triclosan solution	1.25
Water	25.50

100.00%

The triclosan solution replaces water in the basic mastic formulation.

Example 7

A concentrated cleaning solution is provided with an additive of the present invention to provide a solution with the following composition:

	<u>% by weight</u>
Water	52.5
Broad-spectrum biocide (such as a combination of alkyl dimethyl benzyl ammonium chloride and 2-phenyl-phenol)	30.0
Surfactant	3.0
Emulsifier/Dispersant	2.0
40% active triclosan solution	12.5

1 part of cleaning solution is then diluted with 9 parts water immediately prior to use. The cleaning solution is particularly suitable for washing mouldy walls to kill algae and lichen prior to painting.

Example 8

Investigations were carried out into the efficacy of a paint composition containing a solution of Triclosan comprising the following:

Triclosan	50% w/w
Triglyceride	40-50% w/w
Dispersants/surface tension modifiers	0%-10% w/w

The Triclosan solution was incorporated into solvent-borne, solvent-free and water-borne paint formulations at a level of 1% w/w and had no noticeable effect on physical properties.

The biological activity of Rust-Oleum™ paint compositions containing Triclosan solution of the present invention (“treated”) against *Staphylococcus aureus* ATCC 6538 was compared with various types of Rust-Oleum paint products containing no Triclosan (“untreated”). The biological activity of the coated disc against *Staphylococcus aureus* ATCC 6538 was measured using the halo test, including 24 hours pre-diffusion at 4°C. The results of the halo tests are summarized in Table 1 below:

Table 1

Sample	Zone of inhibition/mm S.aureus	
	Halo	gus
Rust-Oleum/Mathys Paracem (single pack). Untreated.	0.0	-/+
Rust-Oleum/Mathys Paracem (single pack). Treated.	>23.0	-
Rust-Oleum/ Mathys Vernac Satin Clear (single pack). Untreated.	0.0	-/+
Rust-Oleum/Mathys Vernac Satin Clear (single pack). Treated.	18.0	-
Rust-Oleum/Mathys Pegakote (two-pack). Untreated	0.0	-/+
Rust-Oleum/Mathys Pegakote (two pack). Treated.	16.0	-
Rust-Oleum/Mathys Paracem Semigloss (single pack). Untreated	0.0	-/+
Rust-Oleum/Mathys Paracem Semigloss (single pack). Treated	>23.0	-

ATCC = American Type Culture Collection

gus = growth under sample

- = complete suppression of growth

-/+ = slight growth

+/- = moderate growth

+ = no suppression of growth

() = partial inhibition

Additionally, the durability of the biological activity of treated and untreated Rust-Oleum Paracem Semi-gloss and Rust-Oleum Pegakote Twin Pack against

Staphylococcus aureus ATCC 6538 was studied using AATCC test method 100-1998 amended. 40mm discs were cleaned for 5 minutes at 100°C with moist heat and inoculated with 0.2ml of culture in one quarter strength Ringers solution to give $10^5 - 10^6$ colonies per disc. The discs were covered with a sterile cover slip (24mm x 24mm) and stored at 20°C at RH > 90% for 7 days. Recovery in Tryptone Soya Broth containing inactivators. Recovery agar plate count at 48 hours at 37°C.

The results of the durability tests are given in Table 2 below and illustrated in Figures 1 and 2 of the accompanying drawings.

Recoveries of viable bacteria

Sample/Time	S.aureus/samples
Inoculation control	5.6x10 E5
Paracem Semi Gloss Untreated	24 hrs 7.6x10 E4
	72 hrs 3.6x10 E4
	7 days 4.0x10 E3
Paracem Semi Gloss Treated	24 hrs 3.2x10 E3
	72 hrs 7.0x10 E2
	7 days <10
Pegakote Twin Pack Untreated	24 hrs 1.0x10 E5
	72 hrs 1.8x10 E4
	7 days 7.1x10 E3
Pegakote Twin Pack Treated	24 hrs 9.4x10 E4
	72 hrs 3.6x10 E2
	7 days < 10

The above tests show that the paint compositions treated with a Triclosan solution according to the present invention exhibit excellent bacteriostatic and bactericidal activity. They prevent/inhibit the growth of micro-organisms, subject to the conditions of the Halo test and organisms used, and also give a higher rate of 'kill' than the untreated products under the conditions of the modified AATCC 100 test. The antimicrobial used is recognised as demonstrating antibacterial activity against a large range of gram+ve bacteria (as tested using Staphylococcus aureus)

and gram-ve bacteria. For example, Triclosan has been shown to be effective against, inter alia, the Gram-positive bacteria *Bacillus cereus*, *Clostridium botulinum*, *Corynebacterium acnes*, *Diplococcus pneumoniae*, *Enterococcus faecalis*, *Lactobacillus arabinosus*, *Listeria monocytogenes*, *Micrococcus luteus* and *Streptococcus aureus* and to be effective against, inter alia, the Gram-negative bacteria *Aerobacter aerogenes*, *Brucella abortus*, *Escherichia coli*, *Proteus vulgaris*, *Pseudomonas aeruginosa*, and *Salmonella enteritidis*. From the results obtained it is reasonable to conclude that good bacteriostatic and bactericidal activity should be exhibited against a large range of bacteria.

The solution of the present invention may be added to a wide range of aqueous and non-aqueous paints and other coating materials which are applied to a substrate to impart bactericidal and/or antimicrobial properties to the coating on the substrate. The non-granular texture of the solution results in it being more acceptable for inclusion in surface coatings vis-à-vis the Triclosan compositions of the prior art. Furthermore, the solution may be added to the surface coating at any stage, during or post-manufacture of the additive and/or surface coating.

The provision of 0.5% active Triclosan solution in the formulation applied to a substrate appears to ensure that the active ingredient is above the Minimum Inhibitory Concentrations 'mic' for any organism and ensures there is plenty spare to diffuse throughout the coating. This results in the bactericidal and/or antimicrobial properties of the coating being maintained throughout its service life.

CLAIMS

1. An additive comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether, or a derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.
2. An additive as claimed in claim 1 wherein the carrier is at least one selected from the group consisting of a long chain fatty acid, a fatty acid ester or an ester of a glycerol.
3. An additive as claimed in claim 2 wherein the carrier is a triglyceride.
4. An additive as claimed in claim 2 or claim 3 wherein the fatty acids or esters have a chain length of 6 to 20 carbon atoms.
5. An additive as claimed in any one of claims 1 to 4 wherein the fatty acids are saturated.
6. An additive as claimed in claim 1 wherein the carrier is selected from the group consisting of linseed oil, mineral oil, castor oil, soya oil, tall oil fatty acids, C₁₂-C₁₅ alkyl benzoate, MCT oil, 2-ethyl hexyl palmitate, 2-di-ethyl hexyl adipate, triglyceride ester, isodecyl oleate, isopropyl myristate,

isopropyl palmitate, isopropyl stearate, butyl myristate and sodium, potassium and amine soaps of fatty acids.

7. An additive as claimed in claim 6 wherein the carrier is selected from the group consisting of at least one C₁-C₂₀ alkyl ester of a long chain alkyl or aryl carboxylic acid and a glycerol ester.
8. An additive as claimed in claim 7 wherein the long chain alkyl or aryl carboxylic acid is selected from the group consisting of a fatty acid, a dicarboxylic acid and a benzoic acid.
9. An additive as claimed in claim 7 or claim 8 wherein the carrier is a triglyceride or a fatty acid ester having the general basic formula:
$$\text{R-O-CO-C}_X\text{H}_Y$$

wherein R is a branched or straight chain alkyl group having at least 3 carbon atoms; X is 13 to 17 and Y is 27 to 35.
10. An additive as claimed in claim 9 wherein the carrier is the triglyceride MCT oil.
11. An additive as claimed in claim 9 wherein R has 3 to 8 carbon atoms.
12. An additive as claimed in claim 11 wherein the carrier is a fatty acid ester selected from the group consisting of isopropyl myristate, 2-ethyl-hexyl palmitate, butyl myristate, isopropyl stearate and isopropyl palmitate.

13. An additive as claimed in any one of the preceding claims wherein the solution contains a maximum of 60% by weight of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof.
14. An additive as claimed in any one of the preceding claims wherein the solution contains at least 30% by weight of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof.
15. An additive as claimed in claim 14 wherein the solution contains 30-50% by weight of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof.
16. An additive as claimed in claim 15 wherein the solution contains 40% by weight of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof.
17. An additive as claimed in any one of the preceding claims further comprising minor amounts of other active ingredients.
18. An additive as claimed in claim 17 wherein the other active ingredients are dispersants or surface tension modifiers.

19. An additive as claimed in claim 18 wherein the other active ingredients are included in an amount 0 to 10% by weight.
20. An additive as claimed in claim 17 wherein the other active ingredients are additional microbiologically active substances.
21. An additive as claimed in claim 20 wherein the microbiologically active substances are included in an amount 0 to 30% by weight.
22. An additive as claimed in any one of the preceding claims wherein the viscosity of the solution is 1mPa (0.01 Poise) to 2 Pas (20 Poise).
23. The use of an additive as claimed in any one of the preceding claims for imparting bactericidal and/or antimicrobial properties to a material.
24. The use of an additive as claimed in claim 23 wherein the material is a surface coating.
25. The use of an additive as claimed in claim 24 wherein the surface coating is a paint formulation.
26. The use of an additive as claimed in claim 24 wherein the surface coating is a mastic or sealant.

27. The use of an additive as claimed in claim 23 wherein the material is a cleaning solution.
28. A method for imparting bactericidal and/or antimicrobial properties to a material comprising adding to the material 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.
29. A method as claimed in claim 28 wherein the additive is added to a surface coating.
30. A method as claimed in claim 29 wherein the additive is added during the manufacture of the surface coating.
31. A method as claimed in claim 29 wherein the additive is added after manufacture of the surface coating.
32. A method as claimed in claim 29, 30 or 31 wherein the surface coating is a paint formulation.
33. A method as claimed in claim 29, 30 or 31 wherein the surface coating is a sealant or mastic.

34. A method as claimed in claim 28 wherein the material is a cleaning solution.
35. A method as claimed in any one of claims 28 to 34 wherein the solution of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof is present in the material in an amount of less than 5% by volume.
36. A method as claimed in claim 35 wherein the solution of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof is present in the material in an amount 0.1 to 2.5% by volume.
37. A method as claimed in claim 36 wherein the solution of 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof is present in the material in an amount 0.5 to 1.5% by volume.
38. A paint composition comprising a binder and a pigment further comprising 2,4,4'-trichloro-2'-hydroxy diphenyl ether, 4,4'-dichloro-2-hydroxy diphenyl ether or a derivative thereof dissolved in a carrier, the carrier comprising at least one selected from the group consisting of a fatty acid, a fatty acid ester, a fatty acid salt, an alkyl ester of benzoic acid, an alkyl

ester of a dicarboxylic acid and an ester of glycerol or a derivative of any of the aforesaid.