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(54) Titre : FORMULATIONS FONGICIDES A CROISSANCE CRISTALLINE REDUITE
(54) Title: FUNGICIDE FORMULATIONS WITH REDUCED CRYSTAL GROWTH

(57) Abrégé/Abstract:
This invention relates to a composition comprising a compound of formula (I) and an agrochemical active ingredient selected from ADEPIDYN™ (pydiflumetofen), oxathiapiprolin, sedaxane and azoxystrobin; $R_1O[C(R_2)(H)C(R_3)(H)O]_nX$ (I), where R_1 is C_{6-12} alkyl or is C_{6-12} alkenyl; n is from 5 to 50; independently, each $[C(R_2)(H)C(R_3)(H)O]$ unit has both R_2 and R_3 being hydrogen or has one of R_2 and R_3 being hydrogen and the other being phenyl; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has one of R_2 and R_3 being hydrogen and the other being phenyl; and X is hydrogen or is selected from C_{1-4} alkyl; and to use of a compound of formula (I) to reduce particle growth (such as nucleation, crystal growth or Ostwald ripening) of an agrochemical active ingredient.

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Abrégé:

La présente invention concerne une composition comprenant un composé de formule (I) et un principe actif agrochimique choisi parmi l'ADEPIDYN™ (pydiflumétofen), l'oxathiapiproline, le sédaxane et l'azoxystrobine ;
R₁O[C(R₂)(H)C(R₃)(H)O]_nX (I), où R₁ représente un alkyle en C6-12 ou un alcényle en C₆₋₁₂ ; n est compris entre 5 et 50 ; indépendamment, chaque motif [C(R₂)(H)C(R₃)(H)O] comporte à la fois R₂ et R₃ étant un atome d'hydrogène ou a l'un de R₂ et R₃ étant un atome d'hydrogène et l'autre étant un phényle ; à condition qu'au moins un motif [C(R₂)(H)C(R₃)(H)O] ait l'un de R₂ et R₃ étant un atome d'hydrogène et l'autre étant un phényle ; et X est un atome d'hydrogène ou est choisi parmi un alkyle en C₁₋₄ ; et l'utilisation d'un composé de formule (I) pour réduire la croissance de particules (telle que la nucléation, la croissance de cristaux ou le mûrissement d'Ostwald) d'un principe actif agrochimique.

Abstract:

This invention relates to a composition comprising a compound of formula (I) and an agrochemical active ingredient selected from ADEPIDYN™ (pydiflumetofen), oxathiapiprolin, sedaxane and azoxystrobin; R₁O[C(R₂)(H)C(R₃)(H)O]_nX (I), where R₁ is C₆₋₁₂ alkyl or is C₆₋₁₂ alkenyl; n is from 5 to 50; independently, each [C(R₂)(H)C(R₃)(H)O] unit has both R₂ and R₃ being hydrogen or has one of R₂ and R₃ being hydrogen and the other being phenyl; provided that at least one [C(R₂)(H)C(R₃)(H)O] unit has one of R₂ and R₃ being hydrogen and the other being phenyl; and X is hydrogen or is selected from C₁₋₄ alkyl; and to use of a compound of formula (I) to reduce particle growth (such as nucleation, crystal growth or Ostwald ripening) of an agrochemical active ingredient.

FUNGICIDE FORMULATIONS WITH REDUCED CRYSTAL GROWTH

This invention relates to a composition comprising a compound of formula (I) and an agrochemical active ingredient selected from ADEPIDYN™ (pydiflumetofen), oxathiapiprolin, sedaxane and azoxystrobin;

$$R_1O[C(R_2)(H)C(R_3)(H)O]_nX \qquad (I)$$

where R_1 is C_{6-12} alkyl or is C_{6-12} alkenyl;
n is from 5 to 50;
independently, each $[C(R_2)(H)C(R_3)(H)O]$ unit has both R_2 and R_3 being hydrogen or has one of R_2 and R_3 being hydrogen and the other being phenyl; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has one of R_2 and R_3 being hydrogen and the other being phenyl; and X is hydrogen or is selected from C_{1-4} alkyl;
and to use of a compound of formula (I) to reduce particle growth (such as nucleation, crystal growth or Ostwald ripening) of an agrochemical active ingredient.

Agrochemical compositions may undergo undesirable particle growth, such as crystal growth, nucleation or Ostwald ripening, when stored for periods of time (for example, in a distributor's warehouse or in a farmer's store), particularly in situations where there is an oscillating temperature (such as freeze-thaw cycling). Such behaviour may lead to adverse performance of the agrochemical compositions (including separation, sedimentation, caking within an agrochemical concentrate; blocking of spray nozzles during attempted application of the agrochemical, either in concentrated form or in a diluted form; and poor biological performance of the agrochemical, possibly due to limited availability of the agrochemical to its intended biological site due to large particle sizes, which have a smaller surface are to volume ratio than corresponding smaller particles and hence a reduced bioavailability).

Surprisingly, it has now been found that certain ethylene oxide-styrene oxide copolymer monoalkyl ether polymers may reduce or overcome the crystal growth behaviour and hence the associated physical and biological problems.

Certain compositions comprising an ethylene oxide-styrene oxide copolymer monoalkyl ether polymer and particular agrochemical active ingredients are novel.

Compounds of formula (I) are known only as wetting agents; not as crystal growth inhibitors.

Therefore, the present invention provides a composition comprising a liquid continuous phase, a compound of formula (I) and an agrochemical active ingredient selected from pydiflumetofen, oxathiapiprolin, sedaxane and azoxystrobin;

$$R_1O[C(R_2)(H)C(R_3)(H)O]_nX \qquad (I)$$

where R_1 is C_{6-12} alkyl or is C_{6-12} alkenyl;

n is from 5 to 50;

independently, each $[C(R_2)(H)C(R_3)(H)O]$ unit has both R_2 and R_3 being hydrogen or has one of R_2 and R_3 being hydrogen and the other being phenyl; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has one of R_2 and R_3 being hydrogen and the other being phenyl; and X is hydrogen or is selected from C_{1-4} alkyl.

This means that each unit in the moiety $[C(R_2)(H)C(R_3)(H)O]_n$ has the formula $[C(R_2)(H)C(R_3)(H)O]$ but each unit is independently selected from the following options: R_2 is hydrogen and R_3 is hydrogen; or R_2 is hydrogen and R_3 is phenyl; or R_2 is phenyl and R_3 is hydrogen; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has R_2 is hydrogen and R_3 is phenyl; or R_2 is hydrogen and R_3 is phenyl.

The moiety $[C(R_2)(H)C(R_3)(H)O]_n$ is a random co-polymer; or a block co-polymer.

Preferably n is from 5 to 40 (more preferably from 5 to 20).

Preferably X is hydrogen.

Alkyl and alkenyl chains may be linear or branched.

Suitably R_1 is C_{6-12} alkyl; more suitably R_1 is C_{8-10} alkyl; even more suitably R_1 is C_8 alkyl.

Suitably the compound of formula (I) is a compound of formula (Ia)

$$R_1O[C(R_4)(H)C(R_5)(H)O]_r[CH_2CH_2O]_sX \qquad (Ia)$$

where, independently, in each $C(R_4)(H)C(R_5)(H)O$ unit R_4 is hydrogen and R_5 is phenyl; or R_4 is phenyl and R_5 is hydrogen; r is from 1 to 25; and s is from 1 to 25; and R_1 is as above or below.

Preferably r is from 1 to 10 (more preferably from 3 to 7).

Preferably s is from 1 to 10 (more preferably from 3 to 7).

Suitably n is an average (mean, median or modal) value.

In another aspect, the present invention provides a novel use of a compound of formula (I) as defined above or below to reduce particle growth (such as nucleation, crystal

growth or Ostwald ripening) in a composition comprising a liquid continuous phase and an agrochemical active ingredient.

The composition may be a solution of an agrochemical (such as a soluble concentrate (SL) or an emulsifiable concentrate (EC)); a suspension of a solid (at room temperature) agrochemical in a liquid continuous phase (either water (SC) or oil (OD)); an emulsion where droplets comprising an agrochemical are dispersed in a liquid continuous phase (either water (EW) or oil (EO)); or may be a suspoemulsion (SE).

The composition may further comprise a surfactant. It is possible that the presence of a surfactant may increase the likelihood of the nucleation, crystal growth or Ostwald ripening behaviour which the compound of formula (I) is used to reduce. Surfactants are compounds which reduce the surface tension of water. Examples of surfactants are ionic (anionic, cationic or amphoteric) and nonionic surfactants.

The noun "agrochemical" and term "agrochemically active ingredient" are used herein interchangeably, and include herbicides, insecticides, nematicides, molluscicides, fungicides, plant growth regulators and safeners; preferably herbicides, insecticides and fungicides.

An agrochemical, or a salt of an agrochemical, selected from those given below, may be suitable for the present invention.

Suitable herbicides include pinoxaden, bicyclopyrone, mesotrione, fomesafen, tralkoxydim, napropamide, amitraz, propanil, pyrimethanil, dicloran, tecnazene, toclofos methyl, flamprop M, 2,4-D, MCPA, mecoprop, clodinafop-propargyl, cyhalofop-butyl, diclofop methyl, haloxyfop, quizalofop-P, indol-3-ylacetic acid, 1-naphthylacetic acid, isoxaben, tebutam, chlorthal dimethyl, benomyl, benfuresate, dicamba, dichlobenil, benazolin, triazoxide, fluazuron, teflubenzuron, phenmedipham, acetochlor, alachlor, metolachlor, pretilachlor, thenylchlor, alloxydim, butroxydim, clethodim, cyclodim, sethoxydim, tepraloxydim, pendimethalin, dinoterb, bifenox, oxyfluorfen, acifluorfen, fluazifop, S-metolachlor, glyphosate, glufosinate, paraquat, diquat, fluoroglycofen-ethyl, bromoxynil, ioxynil, imazamethabenz-methyl, imazapyr, imazaquin, imazethapyr, imazapic, imazamox, flumioxazin, flumiclorac-pentyl, picloram, amodosulfuron, chlorsulfuron, nicosulfuron, rimsulfuron, triasulfuron, triallate, pebulate, prosulfocarb, molinate, atrazine, simazine, cyanazine, ametryn, prometryn, terbuthylazine, terbutryn, sulcotrione, isoproturon, linuron, fenuron, chlorotoluron, metoxuron, iodosulfuron, mesosulfuron, diflufenican, flufenacet, fluroxypyr, aminopyralid, pyroxsulam, XDE-848 Rinskor and halauxifen-methyl.

Suitable fungicides include isopyrazam, mandipropamid, azoxystrobin, trifloxystrobin, kresoxim methyl, mefenoxam, famoxadone, metominostrobin and picoxystrobin, cyprodanil, carbendazim, thiabendazole, dimethomorph, vinclozolin, iprodione, dithiocarbamate, imazalil, prochloraz, fluquinconazole, epoxiconazole, flutriafol, azaconazole, bitertanol, bromuconazole, cyproconazole, difenoconazole, hexaconazole, paclobutrazole, propiconazole, tebuconazole, triadimefon, trtiticonazole, fenpropimorph, tridemorph, fenpropidin, mancozeb, metiram, chlorothalonil, thiram, ziram, captafol, captan, folpet, fluazinam, flutolanil, carboxin, metalaxyl, bupirimate, ethirimol, dimoxystrobin, fluoxastrobin, orysastrobin, metominostrobin, prothioconazole, adepidyn, bixafen, fludioxinil, fluxapyroxad, prothioconazole, pyraclostrobin, revysol, solatenol and xemium.

Suitable insecticides include thiamethoxam, imidacloprid, acetamiprid, clothianidin, dinotefuran, nitenpyram, fiprinil, abamectin, emamectin, tefluthrin, emamectin benzoate, bendiocarb, carbaryl, fenoxycarb, isoprocarb, pirimicarb, propoxur, xylylcarb, asulam, chlorpropham, endosulfan, heptachlor, tebufenozide, bensultap, diethofencarb, pirimiphos methyl, aldicarb, methomyl, cyprmethrin, bioallethrin, deltamethrin, lambda cyhalothrin, cyhalothrin, cyfluthrin, fenvalerate, imiprothrin, permethrin, halfenprox, chlorantraniliprole, oxamyl, flupyradifurone, sedaxane, inscalis, rynaxypyr, sulfoxaflor and spinetoram.

Suitable plant growth regulators include paclobutrazole, trinexapac-ethyl and 1-methylcyclopropene.

Suitable safeners include benoxacor, cloquintocet-mexyl, cyometrinil, dichlormid, fenchlorazole-ethyl, fenclorim, flurazole, fluxofenim, mefenpyr-diethyl, MG-191, naphthalic anhydride and oxabetrinil.

The various editions of The Pesticide Manual [especially the 14th and 15th editions] also disclose details of agrochemicals, any one of which may suitably be used in the present invention.

Suitably the agrochemical is present in the composition at a concentration of from 0.5% to 50.0% (more suitably from 1.0% to 30%; more suitably from 3.0% to 20.0%; most suitably from 5.0% to 15.0%) by weight.

Suitably the agrochemical is in suspended form in the continuous phase.

Suitably, the agrochemical is ADEPIDYN™ (pydiflumetofen), oxathiapiprolin, sedaxane or azoxystrobin.

Suitably the continuous phase is aqueous.

A further agrochemical may be present in the composition of the present invention.

Suitably the further agrochemical is present in the composition at a concentration of from 0.5% to 50.0% (more suitably from 1.0% to 30%; more suitably from 3.0% to 20.0%; most suitably from 5.0% to 15.0%) by weight.

Suitably the first agrochemical is in suspended form in the continuous phase whilst the further agrochemical is present in an emulsified form.

When more than one agrochemical is present, preferably ADEPIDYN™ (pydiflumetofen) is in suspended form whilst propiconazole is present as an emulsion; or oxathiapiprolin is in suspended form whilst mefenoxam is present as an emulsion. In such a situation, suitably the continuous phase is aqueous and the composition is a suspoemulsion.

Suitably the compound of formula (I) is present in the composition at a concentration of from 0.1% to 10.0% (more suitably from 0.3% to 5.0%; more suitably from 0.5% to 2.0%; most suitably from 0.5% to 1.0%) by weight.

The following examples demonstrate the crystal growth associated with compositions according to the present invention. Unless otherwise stated, all concentrations and ratios are by weight.

Particle size distribution is reported as the number of particles that fall into each of the various size ranges, given as a percentage of the total number of all sizes in the sample of interest.

Hence, DV95 (for instance) reports cumulative data; 95 means up to 95% percent of the total particles are smaller than a given number. For example if DV 95=7μm, it means that 95% of the particles are smaller than 7μm and 5% bigger than 7μm.

Break-Thru™ DA647 and Break-Thru™ DA675 are examples of compounds of formula (I).

Break-Thru™ DA647 is Oxirane, phenyl, polymer with oxirane, monooctyl ether (CAS Number: 83653-00-3); it is also known as TEGO™ XP 11010 (from Degussa).

Break-Thru™ DA675 is Oxirane, 2-phenyl-, polymer with oxirane, mono(3,5,5-trimethylhexyl) ether (CAS Number: 303150-42-7); it is also known as TEGO™ VISCOPLUS 3030 and TEGO™ VISCOPLUS 3060 (from Degussa).

Example 1

This example provides data relating to suspoemulsions (SEs) comprising ADEPIDYN™ (pydiflumetofen) suspended in water and propiconazole present as an oil-in-water emulsion. All the SE formulations were prepared and analysed for particle size using conventional processes familiar to those skilled in the art.

Table 1 provides, for each SE, a list of ingredients plus particle size data obtained during Freeze-Thaw (F/T) storage of the SE samples; a Freeze-Thaw cyclic stress test for physical storage of samples was used, whereby every 24hours the storage freezer/oven/temperature was changed from -10°C to 45°C (and then back again 24 hours later). Under such extreme temperature change, stressed material may induce significant particle growth of active ingredients but, as Table 1 shows, this can be mitigated by the presence of a compound of formula (I).

Table 1

Ingredients	ADE-1	ADE-2
Propiconazole	12.5	12.5
Adepidyn™	15	15
1,2,3-Propanetriol	3.7	3.7
Butyl polyalkylene oxide block copolymer	6.0	5.5
Castor oil, ethoxylated	8.0	8.2
Lignosulfonic acid, sodium salt	1.5	0.5
Methyl methacrylate polymer with methacrylic acid and methoxy polyoxyethylene methacrylate	1.0	0.8
Break-Thru™ DA647	0	2.2
Magnesium aluminum silicate dispersion	4.0	4.6
Biocides	0.2	0.2
Thickener	3.9	3.9
pH adjuster	0.2	0.2
Antifoam	0.05	0.05
Water	to 100%	to 100%
Initial Particle Size DV(95) µm	7.2	7.1
After 2weeks F/T cycle: Particle Size DV(95) µm	71.3	14.3
Particle size increase factor	> 10 times	2 times

Conclusion: Formulations ADE-1 and ADE-2 are extremely similar compositions, with the key difference being that ADE-2 contains Break-Thru™ DA647, which is absent in

ADE-1; as Table 1 reveals, surprisingly the presence of Break-Thru™ DA647 significantly reduces the rate of particle growth (crystal growth).

Example 2

This example provides data relating to fungicide suspoemulsions (SEs) comprising OXTP (oxathiapiprolin) suspended in water and MFX (mefenoxam) present as an oil-in-water emulsion. All the SE formulations were prepared and analysed for particle size using conventional processes familiar to those skilled in the art.

Table 2 provides for each SE a list of ingredients plus particle size data obtained during storage of the SE samples.

Table 2. OXTP/MFX Formulation and particle size growth over storage at 54°C

Ingredient	OXTP-1	OXTP-2	OXTP-3	OXTP-4	OXTP-5
OXTP	5	5	5	5	5
MFX	15	15	15	15	15
Pluronic™ PE10400	0.5	0.5	0.5	0.5	0.5
Toximul™ 8320	1	1	1	1	1
Break-Thru™ DA675	0	0.5	1		
Break-Thru™ DA647	0			0.5	1
Xanthan pregel	20	20	20	20	20
Xiamenter Antifoam 1510	0.1	0.1	0.1	0.1	0.1
Water	58.4	57.9	57.4	57.9	57.4
Total	100	100	100	100	100

Initial Particle size Dv50 (µm)	1.69	1.75	1.51	1.77	1.87
Initial Particle size Dv95 (µm)	6.44	5.12	5.01	6.74	6.84
Particle size after storage at 54°C 4wk Dv50 (µm)	5.32	1.94	1.54	2.80	2.87
Particle size after storage at 54°C 4wk Dv95 (µm)	12.07	5.27	5.39	5.62	6.14
Dv50 growth after 54°C 4wk (%)	215.5	10.6	2.0	58.2	53.5

Conclusion: Formulations OXTP-1 to OXTP-5 are extremely similar compositions, with the key difference being that Formulations OXTP-2 to OXTP-5 contain Break-Thru™ DA647 or DA675, which are absent in Formulation OXTP-1; as Table 2 reveals, surprisingly the presence of Break-Thru™ DA647 or DA675 significantly reduces the rate of particle growth (crystal growth).

Example 3

This example provides data relating to fungicide suspension concentrates (SCs) comprising sedaxane suspended in water. All the SC formulations were prepared and analysed for particle size using conventional processes familiar to those skilled in the art. Table 3 provides for each SC a list of ingredients plus particle size data obtained during storage of the SC samples.

Table 3. Sedaxane Formulation compositions and their particle size growth.

Ingredient	SDX-1	SDX-2	SDX-3
Sedaxane	5	5	5
Rhodacal™ DS10 Wetting agent	0.2	0.2	0.2
Toximul™ 8320 dispersant	1		
REAX™ 100M dispersant		1	
Break-Thru™ DA-675 dispersant			1
Rhodiasolve™ Polarclean (water miscible solvent)	10	10	10
Xanthan pregel	25	25	25
Xiamenter™ Antifoam 1510	0.1	0.1	0.1
Water	58.7	58.7	58.7
Total	100	100	100
Initial Particle size Dv50 (µm)	3.3	3.3	3.3
Particle size after storage at 50°C 4weeks. Dv50 (µm)	5.4	7.48	3.33
Dv50 growth after 50°C 4weeks. (%)	62	127	1

Conclusion: Formulations SDX-1 to SDX-3 are extremely similar compositions, with the key difference being that they contain different dispersants; Formulation SDX-3 contains Break-Thru™ DA675, which is absent in Formulations SDX-1 and SDX-2, having been replaced by dispersants of different chemistry; as Table 3 reveals, surprisingly the presence of Break-Thru™ DA675 significantly reduces the rate of particle growth (crystal growth).

Example 4

This example provides data relating to fungicide suspension concentrates (SCs) comprising azoystrobin suspended in water. All the SC formulations were prepared and analysed for particle size using conventional processes familiar to those skilled in the art.

Table 4 provides for each SC a list of ingredients plus particle size data obtained during storage of the SC samples.

Table 4. Azoxystrobin Formulation compositions and their particle size growth over storage at 50°C for 4 weeks.

Ingredient	AZ-1	AZ-2	AZ-3
Azoxystrobin	5	5	5
Morwet™ D425 Wetting agent	0.3	0.3	0.3
Toximul™ 8320 dispersant	1		
REAX™ 100M dispersant		1	
Break-Thru™ DA-675 dispersant			1
Rhodiasolve™ Polarclean (water miscible solvent)	10	10	10
Xanthan pregel	25	25	25
Xiameter™ Antifoam 1510	0.1	0.1	0.1
Water	58.6	58.6	58.6
Total	100	100	100

Initial Particle size, Dv50 (µm)	1.16	1.16	1.16
Particle size after storage at 50°C 4wk, Dv50 (µm)	1.48	1.58	1.23
Dv50 growth after 50°C 4wk (%)	27.5	36.6	6.1

Conclusion: Formulations AZ-1 to AZ-3 are extremely similar compositions, with the key difference being that they contain different dispersants; Formulation AZ-3 contains Break-Thru™ DA675, which is absent in Formulations AZ-1 and AZ-2, having been

replaced by dispersants of different chemistry; as Table 4 reveals, surprisingly the presence of Break-Thru™ DA675 significantly reduces the rate of particle growth (crystal growth).

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CLAIMS

1. A composition comprising a liquid continuous phase, a compound of formula (I) and an agrochemical active ingredient selected from selected from pydiflumetofen, oxathiapiprolin, sedaxane and azoxystrobin;

$$R_1O[C(R_2)(H)C(R_3)(H)O]_nX \qquad (I)$$

where R_1 is C_{6-12} alkyl or is C_{6-12} alkenyl;
n is from 10 to 50;
independently, each $[C(R_2)(H)C(R_3)(H)O]$ unit has both R_2 and R_3 being hydrogen or has one of R_2 and R_3 being hydrogen and the other being phenyl; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has one of R_2 and R_3 being hydrogen and the other being phenyl; and X is hydrogen or is selected from C_{1-4} alkyl.

2. A composition as claimed in claim 1 where X is hydrogen.

3. A composition as claimed in claim 1 or 2 where R_1 is C_{6-12} alkyl.

4. A composition as claimed in 3 where R_1 is C_{8-10} alkyl.

5. A composition as claimed in claim 1, 2, 3 or 4 where n is from 5 to 40.

6. A composition as claimed in claim 1 where the compound of formula (I) is a compound of formula (Ia)

$$R_1O[C(R_4)(H)C(R_5)(H)O]_r[CH_2CH_2O]_sX \qquad (Ia)$$

where, independently, in each $C(R_4)(H)C(R_5)(H)O$ unit R_4 is hydrogen and R_5 is phenyl; or R_4 is phenyl and R_5 is hydrogen; r is from 1 to 25; and s is from 1 to 25.

7. Use of a compound of formula (I) to reduce particle growth in a composition comprising a liquid continuous phase and an agrochemical active ingredient

$$R_1O[C(R_2)(H)C(R_3)(H)O]_nX \qquad (I)$$

where R_1 is C_{6-12} alkyl or is C_{6-12} alkenyl;
n is from 5 to 50;
independently, each $[C(R_2)(H)C(R_3)(H)O]$ unit has both R_2 and R_3 being hydrogen or has one of R_2 and R_3 being hydrogen and the other being phenyl; provided that at least one $[C(R_2)(H)C(R_3)(H)O]$ unit has one of R_2 and R_3 being hydrogen and the other being phenyl; and X is hydrogen or is selected from C_{1-4} alkyl.

8. Use as claimed in claim 7 where X is hydrogen.

9. Use as claimed in claim 7 or 8 where R_1 is C_{6-12} alkyl.

10. Use as claimed in 9 where R_1 is C_{8-10} alkyl.

11. Use as claimed in claim 7, 8, 9 or 10 where n is from 5 to 40.

12. Use as claimed in claim 7 where the compound of formula (I) is a compound of formula (Ia)

$$R_1O[C(R_4)(H)C(R_5)(H)O]_r[CH_2CH_2O]_sX \qquad (Ia)$$

where, independently, in each $C(R_4)(H)C(R_5)(H)O$ unit R_4 is hydrogen and R_5 is phenyl; or R_4 is phenyl and R_5 is hydrogen; r is from 1 to 25; and s is from 1 to 25.