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(21) International Application Number: PCT/US92/00191 (22) International Filing Date: 13 January 1992 (13.01.92) (30) Priority data: 91870007.1 16 January 1991 (16.01.91) EP (34) Countries for which the regional or international application was filed: AT et al. (71) Applicant (for all designated States except US): THE PROCTER & GAMBLE COMPANY [US/US]; One Procter & Gamble Plaza, Cincinnati, OH 45202 (US). (72) Inventor; and (75) Inventor/Applicant (for US only) : DE CUPERE, Marcel, Joseph, Jean [BE/BE]; Op de Vliet 14, B-3010 Kessel-Lo (BE).		(74) Agent: REED, T., David; The Procter & Gamble Company, Ivorydale Technical Ctr., 5299 Spring Grove Ave., Cincinnati, OH 45217-1087 (US). (81) Designated States: CA, JP, US. Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: FOAM CONTROL AGENTS IN GRANULAR FORM (57) Abstract <p>The present invention provides stable, free-flowing foam-control agent in granular form, comprising a silicone antifoam compound, a carrier material, an organic coating material, characterized in, that they contain glycerol, at a weight ratio with the silicone antifoam compound of 1:4 to 2:1. The present invention also provides detergent compositions containing said foam control agent.</p>		

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FOAM CONTROL AGENTS
IN GRANULAR FORM

Technical Field

The present invention is concerned with foam control agents in granular form and with detergent compositions containing such agents.

Background

It has become common practice in the detergent industry to include in detergent compositions materials which are intended to control the amount of foam produced during a washing cycle in a washing machine.

Indeed, excessive sudsing has been found to interfere negatively with the action of the wash liquor upon the fabrics.

Silicone antifoam agents have been found to be particularly effective species, and efforts have been directed to increase the stability of such

agents upon prolonged storage of the detergent compositions containing them.

In GB 1,492,339, it has been proposed to coat silicone antifoam agents with an organic material which is a water soluble or water dispersible, substantially non-surface active, detergent-impermeable material e.g. gelatin, agar or certain reaction products of tallow alcohol and ethylene oxide.

In EP-A-210 721, it has been proposed to coat silicone antifoam agents with water-insoluble fatty acid or fatty alcohols, and in EP-A-210 731 it has been disclosed to coat silicone antifoam agents with a monoester of glycerol and a fatty acid.

While such antifoam agents in granulate form appear to exhibit desirable stability upon storage, when incorporated in detergent compositions, it has been found that such antifoam granulates were not ideally suited for an easy incorporation into detergent compositions in powder form, such as dry-mixing.

It is thus the object of the present invention to provide storage-stable foam controlling agents, in the form of granulates which are free flowing and easily processable into a detergent composition.

SUMMARY OF THE INVENTION

The present invention provides a stable, free-flowing foam-control agent in granular form for inclusion in a detergent composition in powder form, comprising a silicone antifoam compound, a carrier material, preferably starch, an organic

coating material, characterized in, that it contains glycerol, at a weight ratio with the silicone antifoam compound of 1:4 to 2:1.

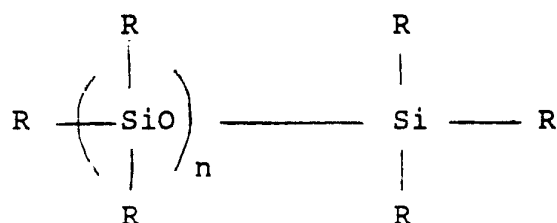
The present invention also provides detergent compositions containing a surface-active agent, said foam control agent, and optionally other detergent ingredients.

DETAILED DESCRIPTION OF THE INVENTION

The silicone antifoam compound

The silicone antifoam compound is present in the foam-control agents of the invention, at levels of from 5% to 20% by weight, preferably 8% to 15% by weight.

In industrial practice, the term "silicone" has become a generic term which encompasses a variety of relatively high-molecular-weight polymers containing siloxane units and hydrocarbyl groups of various types. Generally, the silicone suds-controllers can be described as siloxanes having the general structure :



where each R independently can be an alkyl or an aryl radical. Examples of such substituents are methyl, ethyl, propyl, isobutyl, and phenyl. Preferred polydiorganosiloxanes are polydimethylsiloxanes having trimethylsilyl endblocking units and having a viscosity at 25°C of from $5 \times 10^{-5} \text{ m}^2/\text{s}$ to $0.1 \text{ m}^2/\text{s}$ i.e. a value of n

in the range 40 to 1500. These are preferred because of their ready availability and their relatively low cost.

A preferred type of silicone antifoam agent useful in the compositions herein comprises a mixture of an alkylated siloxane of the type hereinabove disclosed and solid silica.

The solid silica can be a fumed silica, a precipitated silica or a silica made by the gelformation technique. The silica particles suitably have an average particle size of from 0.1 to 50 μ m, preferably from 1 to 20 μ m and a surface area of at least 50m²/g. These silica particles can be rendered hydrophobic by treating them with dialkylsilyl groups and/or trialkylsilyl groups either bonded directly onto the silica or by means of a silicone resin. It is preferred to employ a silica the particles of which have been rendered hydrophobic with dimethyl and/or trimethyl silyl groups. Silicone antifoams employed in a foam control agent according to the invention suitably have an amount of silica in the range of 1 to 30% (more preferably 2.0 to 15%) by weight of the total weight of the silicone antifoam resulting in silicone antifoams having an average viscosity in the range of from 2×10^{-4} m²/s to 1m²/s. Preferred silicone antifoams may have a viscosity in the range of from 5×10^{-3} m²/s to 0.1m²/s. Particularly suitable are silicone antifoams with a viscosity of 2×10^{-2} m²/s or 4.5×10^{-2} m²/s.

The carrier material

The carrier material for use herein provides the solid basis on which the silicone antifoam agent, the organic coating material, and the

glycerol are deposited during manufacture; the carrier material must therefore be in the form of solid particles, which are compatible with detergent ingredients, are water-soluble or water-dispersible to facilitate the dispersion of the silicone antifoam agent in the aqueous liquor during the wash cycle, and are capable to absorb or adsorb the silicone antifoam agent.

The carrier material is present in the present foam control agents at levels of from 40% to 80% by weight, preferably 55% to 65% by weight.

A preferred carrier material for use in foam control agents according to the invention is starch, native or chemically modified both hot or cold gelling in water; other suitable materials are kieselguhr and Fuller's earth.

It is also possible to choose carrier particles for use herein, among ingredients which themselves play an active role in the laundering or washing process. Examples of such materials are zeolites, sodium tripolyphosphate (STPP) particles, sodium sulphate, sodium carbonate, carboxymethylcellulose and clay minerals.

The organic coating material

The organic coating material is present at levels of from 5 to 30% of the antifoam agents according to the invention, preferably from 8% to 22% by weight.

Organic coating material suitable for use in the granules of the present invention, can be water soluble/dispersible species, water-insoluble species, or a mixture thereof.

Water-soluble or water-dispersible carrier materials must be impermeable to detergent and must be substantially non-surface active. By substantially non-surface active is meant that the carrier material, itself, does not interact with the silicone material in such fashion that it is emulsified or otherwise excessively dispersed throughout an aqueous medium, rather than at the air/water interface.

A wide variety of carrier materials having the requisite solubility/dispersibility characteristics and the essential features of being non-surface active and detergent-impermeable are known. For example, the high-molecular-weight Carbowaxes ("Carbowax" is a trade mark) which have substantially not surface-active characteristics are useful herein. Examples of this type of material include the polyethyleneglycols having a molecular weight of from about 1,500 to about 10,000, especially about 4,000. Highly ethoxylated fatty alcohols such as tallow alcohol condensed with about 25 molar proportions of ethylene oxide are useful herein. Other alcohol condensates containing extremely high ethoxylate proportions (ca. 80) are also useful herein. Such high ethoxylates apparently lack sufficient surface-active characteristics to interact or otherwise interfere with the desired suds-control properties of the silicone agents herein. A particularly preferred ethoxylated carrier material herein is tallow alcohol condensed with about 80 molar proportions of ethylene oxide, and abbreviated TAE₈₀.

A variety of other materials useful as water-soluble/dispersible coating agents herein may also be mentioned : gelatin; agar; gum arabic; and various algae-derived gels.

Water-insoluble organic materials suitable for use in a foam control agent according to the invention are water-insoluble fatty acids, fatty alcohols, fatty acid esters and mixtures thereof. Fatty acids or fatty alcohols are species which have from 10 to 20 carbon in their alkyl chain.

Suitable fatty acids are saturated or unsaturated and can be obtained from natural sources such as, for example, plant or animal esters (e.g. palm oil, coconut oil, babassu oil, safflower oil, taloil, castor oil, tallow and fish oils, grase, and mixtures thereof) or can be synthetically prepared for example via the oxidation of petroleum or by hydrogenation of carbon monoxide via the Fisher-Tropsch process. Examples of suitable fatty acids for use in the present invention include capric, lauric, myristic, palmitic, stearic, arachidic and behenic acid.

Preferred is stearic acid. Also preferred is tallow (C₁₆-C₁₈) fatty acid. Alcohols derived from the above-mentioned fatty acids are suitable for the foam control agents herein, stearic alcohol and tallow (C₁₆-C₁₈) alcohol being preferred.

Esters of the above-mentioned fatty acids with C₁₋₃ alcohols are also suitable, such as ethyl myristate, ethyl stearate, methyl palmitate, and glycerol mono stearate, this latter species being especially preferred.

Among the above water-insoluble fatty materials, stearyl acid and stearyl alcohol are especially preferred. These materials are preferred because of their good performance, easy availability and suitable melting point. The melting points of stearic acid and stearyl

alcohol are 71.5 and 59.4°C respectively at which temperatures they are insoluble in water. These materials do not appear to interfere with the effectiveness of the silicone antifoam when it is released into the washing liquor.

Mixtures of the above water-soluble/dispersible coating agents and of the above water-insoluble organic materials herein are also suitable. A preferred mixture is stearyl alcohol and TAE₈₀.

The amount of organic coating material employed in the foam control agents herein should be no less than 1 part organic coating material per part of silicone antifoam.. It is preferred to employ the organic coating material and the silicone antifoam in a weight ratio in the range 5:1 to 1:1, more preferably in the range 4:1 to 2:1.

The glycerol is a key ingredient of the foam control agents herein; it is believed to strengthen the adsorption of silicone antifoam compound to the carrier particles. Glycerol is present at a weight ratio of 1:4 to 2:1 with the silicone antifoam agent, and at levels of from 2 to 20%, preferably 4% to 10% of the foam control agents herein.

Optional ingredients

The foam control agents may comprise, in addition to the essential ingredients described hereinabove, optional ingredients, such as waxes, and anti-caking agents.

Preferred waxes are of mineral origin, especially those derived from petroleum, including microcrystalline and oxidised microcrystalline petroleum waxes and paraffin

waxes. However synthetic waxes, or natural waxes such as Montan waxes, beeswax, candelilla or carnauba waxes may also be used as may a mixture of any of these waxes. Whatever wax is used it is preferred that its melting point is between 35° and 70°C so that it is readily liquifiable.

Suitable anti-caking agents include alkoxylated nonionic surfactants such as described hereinafter.

Making process for the anti-foam agent
of the invention

The process is as follows :

The silicone antifoam compound and the glycerol, at a weight ratio such as presently claimed, are mixed together and the mixture is added to the carrier material, which is preferably starch; the mixing then takes place in an Eirich mixer. During the mixing, the temperature raises due to the agitation. When the temperature reaches a value in the range of from 30 to 35°C, the organic coating material, is added in the molten state (e.g. at 85°C, if stearyl alcohol), and the mixing is continued until approximately 5 minutes after the melting temperature of the organic coating agent is reached (56 to 58°C for stearyl alcohol).

Granulation has then taken place, and the granules are cooled to room temperature in a fluid bed.

Detergent compositions

In another embodiment of the present invention, it is herewith provided a detergent composition

in powder form, comprising a surface-active agent and a foam control agent such as described above. The amount of foam control agents is from 0.25 to 10%, preferably from 0.5 to 3 by weight of the total detergent composition.

Suitable surface-active agents are of the anionic, cationic, non-ionic or amphoteric type, or mixtures thereof. Suitable anionic organic detergent surfactants include alkali metal soaps of higher fatty acids, alkyl aryl sulphonates, for example sodium dodecyl benzene sulphonate, long chain (fatty) alcohol sulphates, olefine sulphates and sulphonates, sulphated monoglycerides, sulphated ethers, sulphosuccinates, alkane sulphonates, phosphate esters, alkyl isothionates, sucrose esters and fluorosurfactants. Suitable cationic organic detergent surfactants include alkyl-amine salts, quaternary ammonium salts, sulphonium salts and phosphonium salts. Suitable non-ionic organic surfactants include condensates of ethylene oxide with a long chain (fatty) alcohol or fatty acid, for example C₁₄₋₁₅ alcohol, condensed with 7 moles of ethylene oxide (Dobanol[®] 45-7), condensates of ethylene oxide with an amine or an amide, condensation products of ethylene and propylene oxides, fatty acid alkylol amides and fatty amine oxides. Suitable amphoteric organic detergent surfactants include imidazoline compounds, alkylaminoacid salts and betaines.

The detergent compositions herein preferably also contain a builder, which is preferably a non-phosphate species; accordingly, the builder herein preferably is selected from aluminosilicate ion exchangers (zeolites), and water-soluble monomeric or oligomeric carboxylate chelating agents such as citrates, succinates,

oxydisuccinates, as well as mixtures of the above species.

Other suitable builder materials include alkali metal carbonates, bicarbonates and silicates, organic phosphonates, amino polyalkylene phosphonates and amino polycarboxylates, ethylene diamine tetraacetic acid and nitrilotriacetic acid. Other suitable water-soluble organic salts are the homo- or co-polymeric polycarboxylic acids or their salts in which the polycarboxylic acid comprises at least two carboxyl radicals separated from each other by not more than two carbon atoms. Polymers of this type are disclosed in GB-A-1,596,756. Examples of such salts are polyacrylates of MW 2000-5000 and their copolymers with maleic anhydride, such copolymers having a molecular weight of from 20,000 to 70,000, especially about 40,000.

Other ingredients which typically form part of a detergent composition in powder form include bleaching agents, such as sodium perborate and percarbonate, bleach activators, anti redeposition agents such as carboxymethyl cellulase, enzymes, brighteners, fabric softening clays, perfumes, dyes, pigments.

Example

A foam-control agent in granular form is produced, having the following composition :

Example I

<u>ingredients</u>	<u>percent by weight</u>
Silicone antifoam compound*	12%
Glycerol	12%
Stearyl Alcohol	18%
Starch	58%

* Polydimethyl siloxane/fumed silica
X2/3419 from Dow Corning

A quantity of 3.4 kg of the above foam control agent in granular form is produced, starting from 0.4 kg silicone antifoam agent, 0.4 kg glycerol, 2 kg starch, and 0.6 kg stearyl alcohol in molten state. The process is described hereinabove.

The above granulates are crispy and free flowing; they are incorporated in a detergent composition in powder form, by dry-mixing.

Thanks to the quality of the granulates, this incorporation into the detergent composition does not raise any processing difficulty.

Example II

Foam control agents according to the invention can be prepared, wherein tallow fatty acid is used instead of stearyl alcohol; such a foam control agent has been prepared, according to the following composition :

<u>Ingredients</u>	<u>percent by weight</u>
Silicone antifoam compound*	12%
Glycerol	6%
Tallow fatty acid	15%
Starch	67%

Typical detergent compositions in powder form in which the present antifoam agents in granular form are included have the following formulae :

<u>(all levels in % by weight)</u>		
<u>Ingredients</u>	<u>Ex. III</u>	<u>Ex. IV</u>
LAS	8.00	6.50
TAS	2.50	3.35
FA45E7	5.00	2.00
Na citrate/citric acid	----	12.00
Zeolite 4A	24.00	16.00
Copolymer AA/MA	5.00	3.50
Phosphonate	0.60	----
EDTA	0.25	0.30
Na carbonate	17.00	10.00
Silicate (R=2)	2.00	2.50
CMC	0.50	----
Clay	----	8.50
PB1	13.00	----
PB4	----	12.00
TAED	5.70	3.20
Enzyme	1.70	1.70
Foam control agent	0.70	0.80
Minors + water	balance to 100%	

The abbreviations for the individual ingredients have the following meaning :

LAS : sodium salt of linear dodecyl benzene sulfonate

TAS : sodium salt of tallow alcohol sulfate

FA45E7 : fatty alcohol (C₁₄-C₁₅) ethoxylated with
about 7 moles of ethylene oxide

Clay : smectite clay

Zeolite 4A : sodium salt of zeolite 4A with average
particle size between 1-10 micrometer

Copolymer AA/MA : copolymer of acrylic acid and maleic acid

CMC : carboxymethylcellulose

Phosphonate : sodium salt of ethylenediamine tetramethylene
phosphonic acid

EDTA : sodium salt of ethylenediamine tetra acetate

PB1 : NaBO₂.H₂O₂ (Sodium Perborate Monohydrate)

PB4 : NaBO₂.H₂O₂.3H₂O (Sodium Perborate Tetrahydrate)

TAED : tetra acetyl ethylene diamine

CLAIMS

1. A stable, free-flowing foam-control agent in granular form for inclusion in a detergent composition in powder form, comprising a silicone antifoam compound, a carrier material, an organic coating material, characterized in, that it contains glycerol, at a weight ratio with the silicone antifoam compound of 1:4 to 2:1.

2. A foam control-agent according to claim 1 wherein the silicone antifoam compound comprises a polydiorganosiloxane and solid silica.

3. A foam-control agent according to claims 1-2 wherein the carrier material is starch.

4. A foam-control agent according to claims 1-3 wherein the organic coating agent is selected from water-soluble or water-dispersible, substantially non-surface active, detergent impermeable materials, from water-insoluble fatty acids, and/or fatty alcohols and/or fatty acid esters, and from mixtures thereof.

5. A foam-control agent according to claim 4 wherein the water-soluble/dispersible, organic coating agent is selected from highly ethoxylated fatty alcohols, condensed with about 25 molar to 80 molar proportions of ethylene oxide.

6. A foam-control agent according to claim 4 wherein the water-insoluble fatty acid and/or fatty alcohols have a carbon chain length of from 12 to 20 carbon atoms and a melting point in the range of from 45°C to 80°C.

7. A foam-control agent according to claims 5 and 6 wherein the organic coating agent is selected from stearic acid, stearyl alcohol, tallow fatty acid, tallow fatty alcohol, tallow alcohol condensed with about 80 molar proportions of ethylene oxide, and mixtures thereof.

8. A foam-control agent according to claims 1-7 which contains 5 to 20% of silicone antifoam agent, from 40% to 80% of carrier material, from 5 to 30% of organic coating material, and from 2 to 20% of glycerol.

9. A method of making a foam-control agent according to claim 1, wherein the silicone anti-foam agent and glycerol are mixed together and added to the carrier material, the organic coating agent is then added, and mixing/granulation follows.

10. A detergent composition in powder form comprising a surface-active agent, and a foam-control agent according to any of claims 1-8.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/US92/00191

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) * According to International Patent Classification (IPC) or to both National Classification and IPC IPC ⁵ C11D 3/08 C11D 9/60 C11D 13/14 US CL 252/89.1, 252/174.12, 174.15, 174														
II. FIELDS SEARCHED <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Minimum Documentation Searched *</div> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 25%; text-align: left; border-bottom: 1px solid black;">Classification System</th> <th colspan="4" style="text-align: left; border-bottom: 1px solid black;">Classification Symbols</th> </tr> <tr> <td style="border-right: 1px solid black; padding: 5px;">US</td> <td style="padding: 5px;">252/89.1</td> <td style="padding: 5px;">252/174.13</td> <td style="padding: 5px;">252/174.15</td> <td style="padding: 5px;">252/174</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Documentation Searched other than Minimum Documentation to the extent that such documents are included in the fields searched *</div>					Classification System	Classification Symbols				US	252/89.1	252/174.13	252/174.15	252/174
Classification System	Classification Symbols													
US	252/89.1	252/174.13	252/174.15	252/174										
III. DOCUMENTS CONSIDERED TO BE RELEVANT *														
Category *	Citation of Document, ** with indication, where appropriate, of the relevant passages †	Relevant to Claim No. ‡												
X Y	EP, A, 0210731 (Burrill) 04 February 1987 See Page 6, line 2, Page 5, lines 16-18 and Page 7, lines 25-32.	1-2, 4-6, 9-10 1-10												
A	EP, A, 0206522 (Foret) 30 December 1986 See the entire document	1-10												
Y	US, A, 4806266 (Burrill) 21 February 1989 See Col. 3, lines 4-50, Col. 4, lines 15-25 and Col. 2, lines 60-65.	1-10												
A	US, A, 4652392 (Bagiowski et al) 24 March 1987	1-10												
P,Y	EP, A, 0414221 (Nebashi) 27 February 1991 See Page 3, lines 10-59.	1-10												
A	US, A, 4,447,349 (Tai) 08 May 1984 See Col. 2, lines 35-39.	4-7												
A	US, A, 4,451,387 (Tai) 29 May 1984 See Col. 1, lines 35-40 and 50-60, Col. 2, lines 62-69	1-10												
<div style="display: flex; justify-content: space-between;"> <div style="width: 48%;"> <p>* Special categories of cited documents: †</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 48%;"> <p>† "later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention"</p> <p>‡ "document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step"</p> <p>§ "document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art."</p> <p>¶ "document member of the same patent family"</p> </div> </div>														
IV. CERTIFICATION														
Date of the Actual Completion of the International Search 06 May 1992	Date of Mailing of this International Search Report <div style="text-align: center; font-size: 1.2em; font-weight: bold;">17 JUN 1992</div>													
International Searching Authority ISA/US	Signature of Authorized Officer <div style="text-align: center;"> Kery A. Fries </div>													

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET

V ☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE¹

This international search report has not been established in respect of certain claims under Article 17(2) (a) for the following reasons:

1 ☐ Claim numbers _____ because they relate to subject matter ¹² not required to be searched by this Authority, namely:

2 ☐ Claim numbers _____ because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out ¹³, specifically:

3 ☐ Claim numbers _____ because they are dependent claims not drafted in accordance with the second and third sentences of PCT Rule 6.4(a).

VI ☐ OBSERVATIONS WHERE UNITY OF INVENTION IS LACKING¹

This International Searching Authority found multiple inventions in this international application as follows:

1 ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims of the international application.

2 ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims of the international application for which fees were paid, specifically claims:

3 ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claim numbers:

4 ☐ As all searchable claims could be searched without effort justifying an additional fee, the International Searching Authority did not invite payment of any additional fee.

Remarks on Protest

- ☐ The additional search fees were accompanied by applicant's protest.
☐ No protest accompanied the payment of additional search fees.