



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(21) International Application Number: PCT/US98/17926</p> <p>(22) International Filing Date: 28 August 1998 (28.08.98)</p> <p>(30) Priority Data:</p> <table border="0"> <tr> <td>60/057,063</td> <td>29 August 1997 (29.08.97)</td> <td>US</td> </tr> <tr> <td>60/086,969</td> <td>28 May 1998 (28.05.98)</td> <td>US</td> </tr> </table> <p>(71) Applicant (for all designated States except US): MICELL TECHNOLOGIES [US/US]; NCSU Centennial Campus, Suite 3500, 1017 Main Campus Drive, Raleigh, NC 27606 (US).</p> <p>(72) Inventors; and</p> <p>(75) Inventors/Applicants (for US only): DeYOUNG, James, P. [US/US]; 106 Lochridge, Durham, NC 27713 (US). STEWART, Gina, M. [US/US]; 2 Chestnut Bluffs Lane, Durham, NC 27713 (US). LAUBACH, Bernadette, Storey [US/US]; 1311 Valley Run, Durham, NC 27707 (US).</p> <p>(74) Agents: SIBLEY, Kenneth, D. et al.; Myers, Bigel, Sibley & Sajovec, P.A., P.O. Box 37428, Raleigh, NC 27627 (US).</p>		60/057,063	29 August 1997 (29.08.97)	US	60/086,969	28 May 1998 (28.05.98)	US	<p>(81) Designated States: AL, AM, AT, AT (Utility model), AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, CZ (Utility model), DE, DE (Utility model), DK, DK (Utility model), EE, EE (Utility model), ES, FI, FI (Utility model), GB, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (Utility model), SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p>Published</p> <p><i>With international search report.</i></p> <p><i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>
60/057,063	29 August 1997 (29.08.97)	US						
60/086,969	28 May 1998 (28.05.98)	US						
<p>(54) Title: END FUNCTIONALIZED POLYSILOXANE SURFACTANTS IN CARBON DIOXIDE FORMULATIONS</p>								
<p>(57) Abstract</p> <p>A method for dry-cleaning articles such as fabrics and clothing in carbon dioxide comprises contacting an article to be cleaned with a liquid dry-cleaning composition for a time sufficient to clean the fabric. The liquid dry-cleaning composition comprises a mixture of carbon dioxide, water, and an end-functional polysiloxane surfactant. After the contacting step, the article is separated from the liquid dry cleaning composition. The method is preferably carried out at ambient temperature.</p>								

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See also U.S. Patents Nos. 5,683,473 to Jureller et al; U.S. Patent No. 5,683,977 to Jureller et al.; U.S. Patent Nos. 5,676,705 to Jureller et al.

U.S. Patent No. 5,377,705 to Smith et al. describes a precision cleaning system in which a work piece is cleaned with a mixture of CO₂ and a co-solvent. Smith provides an entirely non-aqueous system, stating: "The system is also designed to replace aqueous or semi-aqueous based cleaning processes to eliminate the problems of moisture damage to parts and water disposal" (col. 4 line 68 to col. 5 line 3). Co-solvents that are listed include acetone and ISOPAR™ M (col. 8, lines 19-24). Use in dry cleaning is neither suggested nor disclosed. Indeed, since some water must be present in dry-cleaning, such use is contrary to this system.

In view of the foregoing, there is a continuing need for effective carbon dioxide-based cleaning systems.

Summary of the Invention

A method for cleaning articles such as fabrics and clothing in carbon dioxide comprises contacting an article to be cleaned with a liquid dry cleaning composition for a time sufficient to clean the fabric. The liquid dry-cleaning composition comprises a mixture of carbon dioxide, optionally water, and an end-functional polysiloxane surfactant. In one embodiment an organic co-solvent is also included. After the contacting step, the article is separated from the liquid dry cleaning composition. Preferably, the liquid dry cleaning composition is at ambient temperature, of about 0° C to 30° C.

End-functional polysiloxanes used in carrying out the present invention are represented by the formula X₁-A-X₂, wherein X₁ and X₂ are the end-functional groups or moieties (e.g. as described below) and A is a siloxane group such as polydimethylsiloxane.

As discussed below, the end-functional polysiloxane may be provided in liquid CO₂ as a mixture, the mixture useful as an intermediate for preparing the dry cleaning compositions noted above, or useful in other applications where a surfactant in carbon dioxide is desired (including, but not limited to, electronic cleaning operations such as silicon wafer cleaning, cleaning mechanical parts such as gyroscopes, dry cleaning of fabrics, as wetting agents for CO₂ based systems, and as

dispersing additives for compounds including organic and inorganic compounds dispersed in CO₂, as described in U.S. Patent No. 5,789,505).

Also disclosed is an improved a process which involves contacting liquid or supercritical CO₂ with a mobile CO₂-phobic substance. The improvement comprises
5 lowering the surface tension between said CO₂ and CO₂-phobic substance by adding to the CO₂ an end-functional polysiloxane surfactant.

A further aspect of the present invention is a composition comprising: (a) liquid or supercritical carbon dioxide; and (b) from .001 to 30 percent surfactant, wherein said surfactant is an end-functional polysiloxane surfactant having the
10 formula X₁-A-X₂, wherein X₁ and X₂ are CO₂-phobic groups, and A is a polysiloxane group.

A further aspect of the invention is a composition comprising: (a) liquid or supercritical carbon dioxide; (b) from .001 to 30 percent surfactant, wherein said surfactant is an end-functional polysiloxane surfactant; and (c) from .01 to 50 percent
15 of a CO₂-phobic compound, which CO₂-phobic compound is non-covalently associated with said surfactant.

Detailed Description of the Invention

The terms alkyl or loweralkyl as used herein means C1 to C4 linear or
20 branched, saturated or unsaturated alkyl, including methyl, ethyl, propyl, isopropyl, butyl, *sec*-butyl, and *tert*-butyl.

The term "halo" as used herein means halogen, including fluoro, chloro, bromo, and iodo. Fluoro is preferred.

The term "clean" as used herein refers to any removal of soil, dirt, grime, or
25 other unwanted material, whether partial or complete. The invention may be used to clean nonpolar stains (*i.e.*, those which are at least partially made by nonpolar organic compounds such as oily soils, sebum and the like), polar stains (*i.e.*, hydrophilic stains such as grape juice, coffee and tea stains), compound hydrophobic stains (*i.e.*, stains from materials such as lipstick and candle wax), and particulate soils (*i.e.*, soils
30 containing insoluble solid components such as silicates, carbon black, etc.).

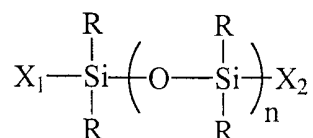
Articles that can be cleaned by the method of the present invention are, in general, garments and fabrics (including woven and non-woven) formed from materials such as cotton, wool, silk, leather, rayon, polyester, acetate, fiberglass, furs,

etc., formed into items such as clothing, work gloves, rags, leather goods (*e.g.*, handbags and brief cases), etc.

“End Functional” Polysiloxane (PSI) materials have specific utility as surfactants in the formulation of CO₂ based cleaning systems. Detergency in non-
 5 aqueous cleaning systems is facilitated by surfactants that increase the quantity and stability of entrained water in the system. End Functional PSI materials are differentiated from other functional PSI materials by the locale and orientation of the functional group (*e.g.*, hydrophilic or lipophilic functional groups; preferably hydrophilic functional groups) being at either (or both) termini of the molecules. The
 10 term “termini” or “terminus” herein refers to the discontinuation or end of dimethyl siloxane repeat units in the molecule. Thus the functional group is typically covalently joined to a dimethyl silyl group, rather than emanating from a methyl siloxane linkage in the backbone of the polymer.

As noted above, end-functional polysiloxane surfactants used in carrying out
 15 the present invention are represented by the formula X₁-A-X₂, wherein X₁ and X₂ are the end-functional groups or moieties (*e.g.* as described below) and A is polysiloxane group such as polydimethylsiloxane, typically having a molecular weight of 100 or 200 to 100,000, 200,000 or 400,000 g/mole. Such compounds are more particularly represented by the formula:

20



wherein:

25 n is from 1 or 2 to 20, 100, 500, 1,000, 5,000, or 10,000. Preferably, n is 1 or 2 to 20, 50 or 100.

At least one, and preferably both, of X₁ and X₂ are CO₂-phobic groups, such as lipophilic or hydrophilic (*e.g.*, anionic, cationic) groups, but are not CO₂-philic groups. The X₁ and X₂ groups may be the same or different. Optionally, but less
 30 preferably, one of X₁ or X₂ may be a CO₂-philic group, such as an alkyl or aryl group as the polysiloxane end terminus.

R is alkyl, aryl, or haloalkyl such as perfluoroalkyl. Lower alkyl, particularly methyl, is preferred, trifluoropropyl is preferred, and phenyl is preferred. Methyl is most preferred.

5 In general, the materials of the invention contain multiple siloxane repeat units that are "CO₂-philic", and CO₂-phobic end-functional groups capable of forming non-covalent associations or positive interactions with desired compounds, such as water, hydrophilic soils, oils, hydrophobic soils, etc.

PSI reactive materials that can be used as precursors for end functional PSI surfactants are silicones with reactive groups that upon reaction with a given substrate yield end functional materials. Reactive groups include but are not limited to; Vinyl, 10 hydride, silanol, alkoxy/polymeric alkoxide, amine, epoxy, carbinol, methacrylate/acrylate, mercapto, acetoxy/chlorine/dimethylamine moieties.

Additional examples of functional end groups include, but are not limited to, hydrophilic end groups such as ethylene glycol, polyethylene glycol, alcohols, 15 alkanolamides, alkanolamines, alkylaryl sulfonates, alkylaryl sulfonic acids, alkylaryl phosphates, alkylphenol ethoxylates, betaines, quarternary amines, sulfates, carbonates, carbonic acids, secondary amines, tertiary amines, aliphatic amines, polyamines, acetylacetate, carbohydrates, anhydrides, malonic esters, alkyl phosphates, glycidyl ethers, and amino acids (including derivatives thereof), etc.; and 20 lipophilic end groups such as linear, branched, and cyclic alkanes, mono and polycyclic aromatic compounds, alkyl substituted aromatic compounds, polypropylene glycol, polypropylene aliphatic and aromatic ethers, fatty acid esters, lanolin, lecithin, lignins (including lignin derivatives), alkyl sulfates, anhydrides, glycidyl ethers, aliphatic amines, and amino acids (including derivatives thereof), etc.

25 An example of an end functional PSI material useful in improving the quantity and stability of water in carbon dioxide and also useful in facilitating detergency is 3-([2-hydroxy-3-diethylamino]propoxy) propyl terminated polydimethylsiloxane. The material has a number average molecular weight of about 200 TO 50,000 g/mole, preferably about 1200 g/mole.

30 The surfactants of the invention can be employed with any carbon-dioxide dry cleaning system, such as described in U.S. Patents No. 5,683,473 to Jureller et al; U.S. Patent No. 5,683,977 to Jureller et al.; U.S. Patent No. 5,676,705 to Jureller et al; and U.S. Patent No. 4,012,194 to Maffei, the disclosures of which applicants specifically

intend to be incorporated herein by reference. Where these prior systems include a surfactant, the surfactants of the invention may be substituted for, or used in combination with, the surfactants described therein.

5 In one particular embodiment, Liquid compositions useful for carrying out the present invention typically comprise:

- (a) from zero or .1 to 10 percent (and when included, more preferably from .1 to 4 percent) water;
- (b) carbon dioxide (to balance; typically at least 30 percent);
- (c) surfactant (preferably from .001, .01, .1 or .5 percent to 5, 10 or 30
10 percent); and
- (d) from zero or .1 to 50 percent (and in one embodiment from 2 or 4 to 30 percent) of an organic co-solvent.

Where a CO₂-phobic substance is included in the composition, it is typically included in an amount of from .01, .1, or .5 to 10, 30, or 50 percent by weight.

15 Percentages herein are expressed as percentages by weight unless otherwise indicated.

The composition is provided in liquid form at ambient, or room, temperature, which will generally be between zero and 50° Centigrade. The composition is held at a pressure that maintains it in liquid form within the specified temperature range. The
20 cleaning step is preferably carried out with the composition at ambient temperature.

The organic co-solvent is, in general, a hydrocarbon co-solvent, including but not limited to alkane, alkene, ether, ester and alcohol cosolvents. Typically the co-solvent is an alkane co-solvent, with C₁₀ to C₂₀ linear, branched, and cyclic alkanes, and mixtures thereof (preferably saturated) currently preferred. The organic co-solvent preferably has a flash point above 140°F, and more preferably has a flash
25 point above 170°F. The organic co-solvent may be a mixture of compounds, such as mixtures of alkanes as given above, or mixtures of one or more alkanes in combination with additional compounds such as one or more alcohols (*e.g.*, from 0 or 0.1 to 5% of a C1 to C15 alcohol (including diols, triols, etc.)).

30 As noted above, other surfactants can be employed in combination with the surfactants of the invention, including surfactants that contain a CO₂-philic group (such as described in U.S. Patents Nos. 5,683,473 to Jureller et al; U.S. Patent No. 5,683,977 to Jureller et al.; U.S. Patent Nos. 5,676,705 to Jureller et al, the disclosures

of which are incorporated herein by reference) linked to a CO₂-phobic group (*e.g.*, a lipophilic group) and surfactants that do not contain a CO₂-philic group (*i.e.*, surfactants that comprise a hydrophilic group linked to a hydrophobic (typically lipophilic) group). A single surfactant may be used, or a combination of surfactants may be used. Numerous surfactants are known to those skilled in the art. *See, e.g.*, McCutcheon's Volume 1: Emulsifiers & Detergents (1995 North American Edition) (MC Publishing Co., 175 Rock Road, Glen Rock, NJ 07452). Examples of the major surfactant types that can be used to carry out the present invention include the: alcohols, alkanolamides, alkanolamines, alkylaryl sulfonates, alkylaryl sulfonic acids, alkylbenzenes, amine acetates, amine oxides, amines, sulfonated amines and amides, betaine derivatives, block polymers, carboxylated alcohol or alkylphenol ethoxylates, carboxylic acids and fatty acids, diphenyl sulfonate derivatives, ethoxylated alcohols, ethoxylated alkylphenols, ethoxylated amines and/or amides, ethoxylated fatty acids, ethoxylated fatty esters and oils, fatty esters, fluorocarbon-based surfactants, glycerol esters, glycol esters, heterocyclic-type products, imidazolines and imidazoline derivatives, isethionates, lanolin-based derivatives, lecithin and lecithin derivatives, lignin and lignin derivatives, maleic or succinic anhydrides, methyl esters, monoglycerides and derivatives, olefin sulfonates, phosphate esters, phosphorous organic derivatives, polyethylene glycols, polymeric (polysaccharides, acrylic acid, and acrylamide) surfactants, propoxylated and ethoxylated fatty acids alcohols or alkyl phenols, protein-based surfactants, quaternary surfactants, sarcosine derivatives, silicone-based surfactants, soaps, sorbitan derivatives, sucrose and glucose esters and derivatives, sulfates and sulfonates of oils and fatty acids, sulfates and sulfonates ethoxylated alkylphenols, sulfates of alcohols, sulfates of ethoxylated alcohols, sulfates of fatty esters, sulfonates of benzene, cumene, toluene and xylene, sulfonates of condensed naphthalenes, sulfonates of dodecyl and tridecylbenzenes, sulfonates of naphthalene and alkyl naphthalene, sulfonates of petroleum, sulfosuccinamates, sulfosuccinates and derivatives, taurates, thio and mercapto derivatives, tridecyl and dodecyl benzene sulfonic acids, etc.

As will be apparent to those skilled in the art, numerous additional ingredients can be included in the dry-cleaning composition, including detergents, bleaches, whiteners, softeners, sizing, starches, enzymes, hydrogen peroxide or a source of hydrogen peroxide, fragrances, etc.

In practice, in a preferred embodiment of the invention, an article to be cleaned and a liquid dry cleaning composition as given above are combined in a closed drum. The liquid dry cleaning composition is preferably provided in an amount so that the closed drum contains both a liquid phase and a vapor phase (that is, so that the drum is not completely filled with the article and the liquid composition). The article is then agitated in the drum, preferably so that the article contacts both the liquid dry cleaning composition and the vapor phase, with the agitation carried out for a time sufficient to clean the fabric. The cleaned article is then removed from the drum. The article may optionally be rinsed (for example, by removing the composition from the drum, adding a rinse solution such as liquid CO₂ (with or without additional ingredients such as water, co-solvent, etc.) to the drum, agitating the article in the rinse solution, removing the rinse solution, and repeating as desired), after the agitating step and before it is removed from the drum. The dry cleaning compositions and the rinse solutions may be removed by any suitable means, including both draining and venting.

Any suitable cleaning apparatus may be employed, including both horizontal drum and vertical drum apparatus. When the drum is a horizontal drum, the agitating step is carried out by simply rotating the drum. When the drum is a vertical drum it typically has an agitator positioned therein, and the agitating step is carried out by moving (*e.g.*, rotating or oscillating) the agitator within the drum. A vapor phase may be provided by imparting sufficient shear forces within the drum to produce cavitation in the liquid dry-cleaning composition. Finally, in an alternate embodiment of the invention, agitation may be imparted by means of jet agitation as described in U.S. Patent No. 5,467,492 to Chao et al., the disclosure of which is incorporated herein by reference. As noted above, the liquid dry cleaning composition is preferably an ambient temperature composition, and the agitating step is preferably carried out at ambient temperature, without the need for associating a heating element with the cleaning apparatus.

In addition, the end-functional polysiloxane surfactants, and liquid or supercritical CO₂ compositions containing the same, can be used for purposes including, but not limited to, electronic cleaning operations such as silicon wafer cleaning, cleaning mechanical parts such as gyroscopes, dry cleaning of fabrics, as wetting agents for CO₂ based systems, and as dispersing additives for compounds

including organic and inorganic compounds dispersed in CO₂, as described in U.S. Patent No. 5,789,505, the disclosure of which is incorporated by reference herein in its entirety.

The present invention is explained in greater detail in the following non-limiting examples.

Example 1

1.032 grams of 3-([2-hydroxy-3-diethylamino]propoxy)propyl terminated polydimethylsiloxane with a number average molecular weight of ~1200 g/mole was added to a 10cc high pressure cell with 100mg of H₂O. Liquid CO₂ was then added to the cell at ambient temperature to a pressure of 2000 psi and stirring was initiated. Over a period of ~2 min the mixture went from dual phase mostly opaque to homogeneous and very slightly hazy. After 10 min the mixture was still homogeneous. The same experiment without added end functional siloxane resulted in a mixture that remained dual phase upon continued stirring.

Example 2

3-([2-hydroxy-3-diethylamino]propoxy) propyl terminated polydimethylsiloxane as described above is synthesized as follows. Starting with epoxypropoxypropyl terminated polydimethylsiloxane with an average molecular weight range of 900-1100 g/mole, the siloxane and a 5 molar excess of diethyl amine are added to a round bottom flask equipped with a reflux condenser. A heating bath is applied to the round bottom flask with a bath temperature of about 78°C and the mixture is refluxed under a static argon head pressure for about 48 hours. The product is isolated by distilling the excess diethyl amine from the polymer and exposing the polymer to a vacuum <1 mm Hg for 12 hours.

The foregoing is illustrative of the present invention, and is not to be construed as limiting thereof. The invention is defined by the following claims, with equivalents of the claims to be included therein.

That which is claimed is:

1. A method for cleaning garments or fabrics in carbon dioxide, comprising:
contacting a garment or fabric article to be cleaned with a liquid dry cleaning
composition for a time sufficient to clean the article, said liquid dry-cleaning
5 composition comprising a mixture of carbon dioxide, water, and a surfactant
wherein said surfactant is an end-functional polysiloxane surfactant; and then
separating the article from the liquid dry cleaning composition.
2. A method according to claim 1, wherein said liquid dry cleaning
10 composition is at a temperature of 0° C to 30° C.
3. A method according to claim 1, wherein said surfactant has the formula X₁-
A-X₂, wherein X₁ and X₂ are CO₂-phobic groups, and A is a polysiloxane group.
- 15 4. A method according to claim 1, wherein said surfactant is 3-([2-hydroxy-
3-diethylamino]propoxy) propyl terminated polydimethylsiloxane.
5. A method according to claim 1, said composition further comprising an
organic co-solvent.
20
6. A liquid dry-cleaning composition, said composition comprising:
(a) from .1 to 10 percent water;
(b) carbon dioxide;
(c) from .1 to 10 percent surfactant, wherein said surfactant is an end-
25 functional polysiloxane surfactant; and
(d) from zero to 50 percent of an organic co-solvent.
7. A liquid dry-cleaning composition according to claim 6, said composition
comprising:
30 (a) from .1 to 4 percent water;
(b) carbon dioxide;
(c) from .5 to 5 percent surfactant; and
(d) from 4 to 30 percent of an organic co-solvent.

8. A liquid cleaning composition according to claim 6, wherein said surfactant is 3-([2-hydroxy-3-diethylamino]propoxy) propyl terminated polydimethylsiloxane.
- 5 9. The compound 3-([2-hydroxy-3-diethylamino]propoxy) propyl terminated polydimethylsiloxane.
- 10 10. The compound according to claim 9 having a number average molecular weight of about 200 to 50,000 g/mole.
- 10 11. The compound according to claim 9 having a number average molecular weight of about 1200 g/mole.
- 15 12. In a process which involves contacting liquid or supercritical CO₂ with a mobile CO₂-phobic substance, the improvement which comprises lowering the surface tension between said CO₂ and CO₂-phobic substance by adding to the CO₂ an end-functional polysiloxane surfactant.
- 20 13. A process according to claim 12, wherein said surfactant is added in an amount from .001 to 30 percent by weight based upon the combined weight of CO₂ and surfactant.
- 25 14. A process according to claim 12, wherein said CO₂ is liquid CO₂.
- 25 15. A process according to claim 12, wherein said surfactant has the formula X₁-A-X₂, wherein X₁ and X₂ are CO₂-phobic groups, and A is a polysiloxane group.
- 30 16. A composition, comprising:
 (a) liquid or supercritical carbon dioxide; and
 (b) from .001 to 30 percent surfactant, wherein said surfactant is an end-functional polysiloxane surfactant having the formula X₁-A-X₂, wherein X₁ and X₂ are CO₂-phobic groups, and A is a polysiloxane group.

17. A composition according to claim 16, wherein said carbon dioxide is liquid carbon dioxide.

18. A composition, comprising:

- 5 (a) liquid or supercritical carbon dioxide;
- (b) from .001 to 30 percent surfactant, wherein said surfactant is an end-functional polysiloxane surfactant;
- (c) from .01 to 50 percent of a CO₂-phobic compound, which CO₂-phobic compound is non-covalently associated with said surfactant.

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19. A composition according to claim 18, wherein said carbon dioxide is liquid carbon dioxide.

20. A composition according to claim 18, said surfactant having the formula
15 X₁-A-X₂, wherein X₁ and X₂ are CO₂-phobic groups, and A is a polysiloxane group

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 98/17926

A. CLASSIFICATION OF SUBJECT MATTER D 06 L 1/04, C 11 D 1/82, C 08 G 77/26		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) D 06 L, C 11 D, C 08 G, B 08 B		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 97/16264 A1 (THE UNIVERISTY OF NORTH CAROLINA AT CHAPEL HILL) 09 May 1997 (09.05.97), claims, examples 25,49-51, 53 (cited in the application). --	1-3, 5- 7, 12- 20
X	WO 96/27704 A1 (UNILEVER N.V.) 12 September 1996 (12.09.96), claims 1,5,9,15, page 29, lines 18-31, page 37, lines 6-8 (cited in the application). ----	1-3, 5- 7, 12- 20
<input type="checkbox"/> Further documents are listed in the continuation of box C.		
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"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family	
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Date of the actual completion of the international search 12 November 1998	Date of mailing of the international search report 22. 12. 98	
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ANHANG

zum internationalen Recherchenbericht über die internationale Patentanmeldung Nr.

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ANNEX

to the International Search Report to the International Patent Application No.

PCT/US 98/17926 SAE 205847

This Annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The Office is in no way liable for these particulars which are given merely for the purpose of information.

ANNEXE

au rapport de recherche international relatif à la demande de brevet international n°

La présente annexe indique les membres de la famille de brevets relatifs aux documents de brevets cités dans le rapport de recherche international visée ci-dessus. Les renseignements fournis sont donnés à titre indicatif et n'engagent pas la responsabilité de l'Office.

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