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(54) **COMPOUNDS AND COMPOSITIONS USEFUL FOR SCAVENGING HYDROGEN SULPHIDE AND SULFHYDRYL-CONTAINING COMPOUNDS**

(57) The present invention relates to a compound obtainable by reaction of at least:

(i) an amino compound comprising at least one primary amine group, this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with

(ii) a polyhydroxy carboxylic acid or an ester thereof; and then with

(iii) formaldehyde.

The present invention also relates to the use of such a compound for scavenging hydrogen sulphide and organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream, and to a composition comprising such a compound and a liquid solvent.

EP 4 279 566 A1

Description

[0001] The present invention relates to novel compounds useful for scavenging hydrogen sulphide and organic compounds comprising at least one sulfhydryl group.

[0002] The present invention also relates to the use of such compounds for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream, in particular in hydrocarbon streams such as crude oils, hydrocarbon fractions derived from the distillation of crude oils, natural gas.

[0003] The present invention also relates to compositions containing such compounds and to a method for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream containing them, comprising contacting said stream with the scavenging compounds of the invention.

PRIOR ART AND BACKGROUND OF THE INVENTION

[0004] Hydrogen sulphide (H_2S) is a colourless and fairly toxic, flammable and corrosive gas which has a characteristic odour at a very low concentration. Hydrogen sulphide dissolves in liquid and gaseous streams such as hydrocarbon and water streams and can also be present in the vapour phase above liquid streams as well as in hydrocarbon gas such as LPG and natural gas. The hydrogen sulphide emissions can be harmful to workers operating in the drilling, production, transport, storage, and processing of such streams. It would therefore be desirable for the workers' comfort and safety to reduce or even eliminate the hydrogen sulphide emissions during the handling of said products.

[0005] Legislation has been in place for years, imposing strict regulations on hydrogen sulphide levels of hydrocarbon streams in pipelines, in storage and shipping containers.

[0006] Similar problems arise with organic compounds comprising a sulfhydryl group (-SH) such as mercaptans R-SH, thiocarboxylic acids $RC(O)SH$, dithiocarboxylic acids $RC(S)SH$, with R denoting a hydrocarbon chain. Such sulfhydryl-containing compounds are very corrosive, and are likely to release hydrogen sulphide.

[0007] A variety of chemical scavengers are available to reduce the concentration of hydrogen sulphide and sulfhydryl-containing compounds in liquid and gaseous streams containing them, in particular aqueous streams and hydrocarbon streams such as gas, crude oils and refined products. Some of the most common methods for treating hydrogen sulphide consist in contacting them with a chemical scavenger such as compounds containing a triazine group, glyoxal, as well as metal-based scavengers. Glyoxal has been used extensively as hydrogen sulphide scavenger but suffers from a major drawback since aqueous glyoxal solutions are highly corrosive and cannot be used for a gas tower application. Triazines have recently become more common chemical scavengers used for treating hydrogen sulphide from hydrocarbon streams. However, many drawbacks are reported that are linked to the use of triazines, including in particular toxicity issues.

[0008] Other hydrogen sulphide scavengers have been developed, and among them scavengers based on oxazolidine, like 3,3'-methylenebis(5-methyloxazolidine), known as MBO. A method for scavenging hydrogen sulphide from sour hydrocarbon substrates has been described in WO 98/02501. MBO presents the advantage of being less toxic and of creating no scales in the conditions where triazine does.

[0009] However, this technology requires an important contact time in order to be efficient in sulphur removal and thus involves injection of higher doses.

[0010] Formulations of MBO with promoters, also named boosters, have been developed to enhance the efficiency of MBO. For example, WO 2017/102693 describes a composition comprising MBO and one or more additive selected among urea, urea derivatives, amino acids, guanidine, guanidine derivatives or 1,2-diols.

[0011] There remains a continuous need for novel solutions for eliminating hydrogen sulphide and other compounds comprising a sulfhydryl group in an efficient, economic and safe manner.

OBJECT OF THE INVENTION

[0012] The Applicant has now discovered that particular novel compounds, as defined below, were particularly efficient for scavenging hydrogen sulphide and unwanted organic compounds comprising at least one sulfhydryl group.

[0013] Such compounds have a high scavenging efficiency, and therefore allow a very efficient removal of hydrogen sulphide and sulfhydryl-containing compounds in a short contact time.

[0014] They have a good solubility in water as well as in organic streams, and therefore can be used for treating both types of streams. Due to this dual solubility, they are also very efficient for treating multi-phase systems such as mixtures of petroleum oil and water which is the common form in which crude oil is extracted from the wellbore.

[0015] Additionally, the compounds used in the present invention are not harmful to human beings and to the environment, they do not give rise to corrosive or other aggressive products, or to solid products which may plug lines or deposit in storage vessels.

[0016] The present invention therefore concerns a compound obtainable by reaction of at least:

(i) an amino compound comprising at least one primary amine group, this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with

(ii) a polyhydroxy carboxylic acid or an ester thereof; and then with
 (iii) formaldehyde.

[0017] The present invention further concerns a method for preparing such a compound.

[0018] The present invention additionally concerns a composition containing at least one liquid solvent and at least one compound as defined above.

[0019] The compound and the composition of the present invention are particularly useful for scavenging hydrogen sulphide and organic compounds comprising at least one sulfhydryl group such as mercaptans, thiocarboxylic acids, dithiocarboxylic acids.

[0020] The present invention also concerns the use of a compound as defined above for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream.

[0021] The present invention further encompasses a method for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream containing them. Such a method comprises contacting said stream with a compound as defined above.

[0022] Other objects, features, aspects and advantages of the invention will become more apparent upon reading the following description and examples.

[0023] In the following, and at least one other indication, the limits of a value range are included within this range, particularly in the expressions "between" and "ranging from ... to ...".

[0024] Moreover, the expressions "at least one" and "at least" used in the present description are respectively equivalent to the expressions "one or more" and "more than or equal to".

[0025] Finally, in a manner known per se, C_N compound or group designates a compound or a group containing in its chemical structure N carbon atoms.

DETAILED DESCRIPTION

The scavenging compound

[0026] The compound of the invention is obtainable by reacting at least the following starting materials:

(i) an amino compound comprising at least one primary amine group (that is to say a group of formula -NH₂), this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with

(ii) a polyhydroxy carboxylic acid or an ester thereof; and then with
 (iii) formaldehyde.

[0027] According to a preferred embodiment, the compound of the present invention is effectively obtained by reacting starting materials (i), (ii) and (iii) as disclosed therein.

[0028] However, any compound which has the same chemical structure as the compound obtained by reacting starting materials (i), (ii) and (iii), but which is obtained from different starting materials (such as for example from chemical equivalents thereof) is intended to be part of the present invention.

[0029] Furthermore, the compound of the invention is obtainable by reacting at least starting materials (i), (ii) and (iii), which means that additional compounds, different from compounds (i), (ii) and (iii) may be involved, such as a quaternizing agent as described below.

[0030] The amino compound (i) is obtained by reacting an acylating agent substituted by a hydrocarbon group and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols.

[0031] The acylating agent is advantageously selected from mono- or poly-carboxylic acids and their derivatives, particularly their ester, amide or anhydride derivatives. The acylating agent is preferably selected from succinic, phthalic and propionic acids and the corresponding anhydrides.

[0032] According to a particularly preferred embodiment, the acylating agent is succinic anhydride.

[0033] The acylating agent is substituted by a hydrocarbon group which can be chosen from any group containing at least 8 carbon atoms wherein a carbon atom is attached directly to the acylating agent.

[0034] Such hydrocarbon groups may also contain heteroatoms such as oxygen, nitrogen, halogens. Such heteroatoms

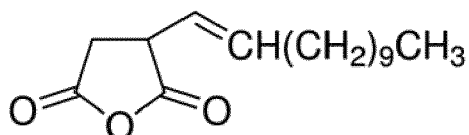
may be inserted into the hydrocarbon main chain, or be part of substituent groups (such as hydroxy groups, amino groups, halogen containing groups or halogen atoms) attached to the hydrocarbon main chain.

[0035] Nevertheless, preference is given to hydrocarbon groups which do not contain such heteroatoms and thus only contain carbon and hydrogen atoms.

[0036] According to a first preferred embodiment, said hydrocarbon group containing at least 8 carbon atoms is chosen from linear or branched, saturated or unsaturated aliphatic C₈-C₂₂ hydrocarbon groups, more preferably from C₈-C₂₂ alkyl or mono or polyunsaturated alkenyl groups.

[0037] Such hydrocarbon groups preferably contain from 8 to 20 carbon atoms, more preferably from 10 to 18 carbon atoms. Examples of hydrocarbon groups substituting the acylating agent include the following groups: n-octyl, n-decyl, n-dodecyl, tetrapropenyl, n-octadecyl, oleyl, octadecyl, dodecenyl.

[0038] A particularly preferred example of acylating agent substituted by a hydrocarbyl group is dodeceny succinic anhydride (DDSA), which corresponds to the following formula:



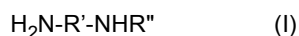
[0039] According to a second preferred embodiment, said hydrocarbon group containing at least 8 carbon atoms is chosen from homo- and co-polymers of mono- and di-olefins having from 2 to 10 carbon atoms, such as for example ethylene, propylene, 1-butene, isobutene, butadiene, isoprene, 1-hexene or 1-octene. Preferably, these olefins are 1-monoolefins.

[0040] In this second embodiment, said hydrocarbon group may contain up to approximately 200 carbon atoms.

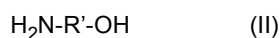
[0041] According to a preferred variant of this embodiment, the hydrocarbon substituent of the acylating agent is a polyisobutene (PIB) group. So-called highly reactive polyisobutenes are most particularly preferred. Highly reactive polyisobutenes means polyisobutenes wherein at least 50% by moles, preferably at least 70% by moles or more, of the terminal olefinic double bonds are of the vinylidene type as described in the document EP0565285. In particular, the preferred PIB are those having more than 80% by moles and up to 100% by moles of terminal vinylidene groups such as described in the document EP1344785.

[0042] Particularly preferred acylating agents substituted by a hydrocarbon group are polyisobutenyl succinic anhydrides (PIBSA). The polyisobutenyl group (PIB group) has, preferably, a number average molecular weight (M_n) between 170 and 2800, for example between 250 and 1500, more preferably between 500 and 1500 and, even more preferably between 500 and 1100. An M_n value range between 700 and 1300 is particularly preferred, for example from 700 to 1000.

[0043] The compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols is preferably chosen from the following amines of formula (I) and (II) below:



and



wherein

R' denotes a C₁ to C₁₀ hydrocarbon chain optionally interrupted by one or more oxygen or nitrogen atoms, preferably

R' denotes a C₁ to C₄ alkylene group or a group or formula -(CH₂-CH₂-O)_n- wherein n is an integer ranging from 1 to 4 and preferably n is 2 or 3, and

R'' denotes hydrogen or a C₁ to C₄ alkyl group.

[0044] Preferred compounds are diamines of formula (I) above, and in particular those wherein R'' denotes hydrogen.

[0045] A particularly preferred compound is ethylenediamine.

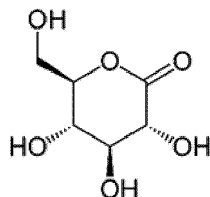
[0046] Further preferred compounds include diamines of formula H₂N-(CH₂-CH₂-O)_n-NH₂, wherein n is an integer ranging from 1 to 4 and preferably n is 2 or 3. Such amines are commercially available under the name Jeffamine[®].

[0047] The polyhydroxy carboxylic acid (ii) is preferably chosen from those containing from 3 to 10 carbon atoms, more preferably from 4 to 8 carbon atoms, and bearing at least two hydroxy (-OH) groups. Particularly preferred compounds (ii) are sugar acids - that is to say monosaccharides bearing a carboxyl group (-COOH)- and esters thereof, including internal esters (lactone forms).

[0048] More particularly preferred compounds are aldonic acids such as glyceric acid, xylonic acid, gluconic acid, and gluconolactone.

[0049] A particularly preferred compound is δ -gluconolactone which corresponds to the following formula:

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[0050] The third compound (iii) is formaldehyde, which is a well-known reagent of formula $H_2C=O$.

[0051] According to a particular embodiment of the invention, the compound the invention is obtainable by further reacting the condensation product of compounds (i), (ii) and (iii) with (iv) a quaternizing agent.

[0052] In this embodiment, the compound of the invention is under the form of a quaternary ammonium salt of the condensation product of compounds (i), (ii) and (iii).

[0053] The quaternizing agent (iv) can advantageously be chosen from dialkyl sulfates, carboxylic acid esters, alkyl halides, benzyl halides, hydrocarbon carbonates, and hydrocarbon epoxides optionally mixed with an acid.

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The method of preparation

[0054] The present invention further concerns a method of preparation of the scavenging compounds of the invention, comprising the following steps:

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(a) a step of condensation of an amino compound (i) comprising at least one primary amine group (that is to say a group of formula $-NH_2$), this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with a polyhydroxy carboxylic acid or an ester thereof (ii); followed by

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(b) a step of condensation of the product of step (a) with formaldehyde (iii).

[0055] The scavenging compounds of the invention are obtained by first (step a) reacting said amino compound (i) comprising at least one primary amine group with said polyhydroxy carboxylic acid or ester thereof (ii). Compounds (i) and (ii) are preferably employed in stoichiometric amounts, of 0.8 to 1.2 moles of compound (ii) per mole of compound (i).

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[0056] Then the resulting condensation product (the product of step a) is reacted with formaldehyde (iii). The amount of formaldehyde employed typically ranges from 3 to 10 moles per mole of said product of step (a).

[0057] Preferred embodiments for specific starting components (i) and (ii) are described above and also apply to the method of the invention.

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[0058] Steps (a) and (b) are carried out in a suitable medium, such as for example an aromatic solvent such as toluene.

[0059] In a preferred embodiment, the method of the invention comprises:

(a) a step of condensation of an amino compound (i) which is obtained by condensing a hydrocarbon-substituted anhydride such as dodeceny succinic anhydride (DDSA) or a polyisobutenyl succinic anhydride (PIBSA) with a diamine of formula (I) above wherein R'' denotes hydrogen; with δ -gluconolactone (ii); followed by

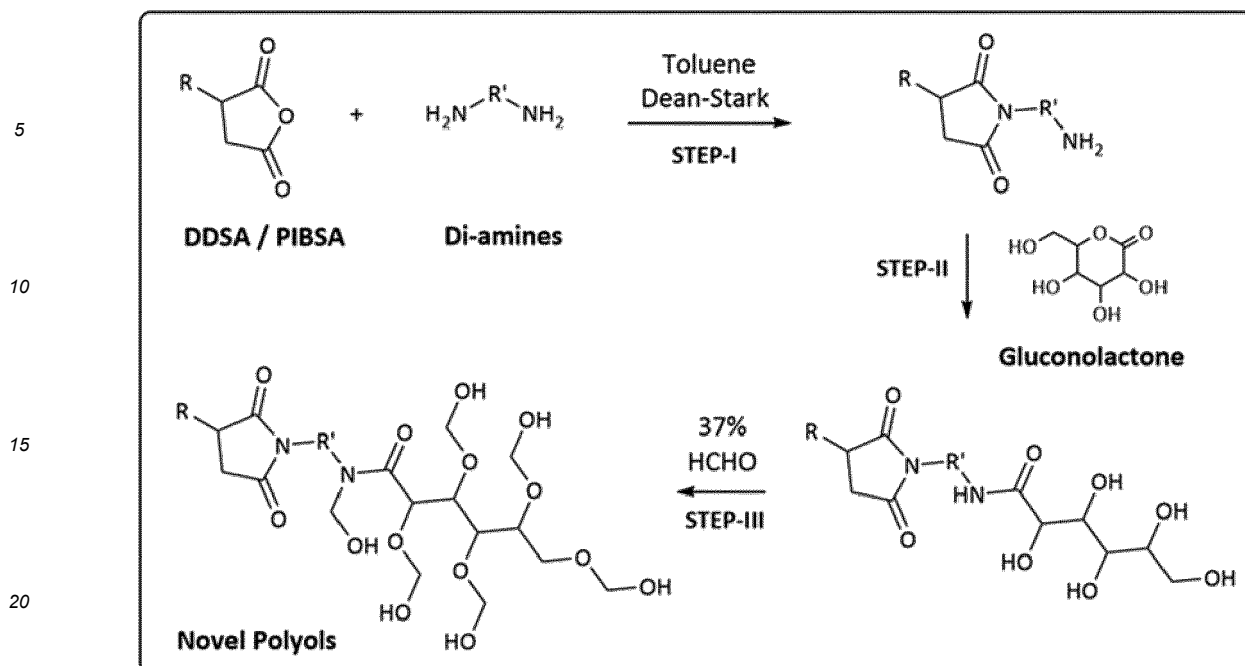
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(b) a step of condensation of the product of step (a) with formaldehyde (iii).

[0060] In this embodiment, the method of the invention can be illustrated by the following reaction scheme:

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25 **[0061]** According to a particular embodiment, the method of preparation according to the invention comprises after step (b) a further step (c) of quaternization of the compound resulting from step (b).

[0062] In this embodiment, the product of step (b) (that is to say, the condensation product of compounds (i), (ii) and (iii)) is reacted with a quaternizing agent (iv).

30 **[0063]** Said quaternizing agent (iv) can advantageously be chosen from dialkyl sulfates, carboxylic acid esters, alkyl halides, benzyl halides, hydrocarbon carbonates, and hydrocarbon epoxides optionally mixed with an acid.

The composition

[0064] The composition of the present invention contains one or more liquid solvent(s), and one or more scavenging compound(s) as defined above.

35 **[0065]** By liquid, it is meant a solvent which is in liquid form at ambient temperature (20°C) and atmospheric pressure (1,013.10⁵ Pa).

[0066] The solvent may be chosen from water and organic solvents.

[0067] According to a preferred embodiment, the solvent is selected from organic liquid solvents.

40 **[0068]** Such solvents may in particular be chosen from poly alkyl ethers, aliphatic hydrocarbons such as alkanes, aromatic solvents such as aromatic hydrocarbons and aromatic hetero-compounds, and mixtures thereof.

[0069] Preferred organic solvents are chosen from aromatic solvents, such as N-methylpyrrolidone, xylene, toluene, benzene; and poly alkyl ethers such as butyl carbitol (diethylene glycol monobutyl ether); as well as mixtures thereof.

45 **[0070]** Other preferred organic solvents include those derived from biomass, such as oils of vegetable origin. A particularly preferred solvent is cashew nutshell liquid, also known as CNSL, which is a widely available vegetable oil derived from cashew nut shell. CNSL can be used as a mixture with any other solvent such as those described herein.

[0071] The choice of the solvent mainly depends on the final use of the composition. When the composition is intended to be used for scavenging hydrogen sulphide and/or sulfhydryl-containing compounds in an aqueous stream, water may be a solvent of choice. On the other side, when the composition is intended to be used for scavenging hydrogen sulphide and/or sulfhydryl-containing compounds in a hydrocarbon stream, an organic solvent will be preferred.

50 **[0072]** When the stream is a mixture of water and hydrocarbons (such as a mixture of crude oil and brine) a solvent having a dual solubility, i.e. a water solubility and a solubility in hydrocarbons, can be preferred. Poly alkyl ethers such as butyl carbitol are preferred solvents since they have such a dual solubility.

[0073] The composition of the invention advantageously contains an amount of solvent ranging from 25 to 95% by weight, preferably from 40 to 90%wt of the composition, more preferably from 50 to 80%wt, relative to the total weight of the composition.

55 **[0074]** The composition advantageously contains said scavenging compound in an amount ranging from 5 to 75% by weight, preferably from 10 to 60% by weight, more preferably from 20 to 50% by weight, relative to the total weight of the composition.

The use

[0075] The compound of the invention as described above is particularly useful for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream.

[0076] Thus, the present invention encompasses the use of a compound as described above for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream.

[0077] According to a preferred embodiment, such compound is used in a composition containing one or more liquid solvents as described previously.

[0078] Thus, the present invention also encompasses the use of a composition as described above for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream.

[0079] The organic compounds comprising at least one sulfhydryl group are especially chosen from mercaptans, thiocarboxylic acids, dithiocarboxylic acids.

[0080] The liquid or gaseous stream may be a monophasic stream such as in particular an aqueous stream or an organic stream, or a multiphasic stream containing both water and hydrocarbons (such as for instance oil/water or oil/water/gas or gas/water).

[0081] According to a preferred embodiment, the compounds of the invention are used for scavenging hydrogen sulphide (H₂S) and mercaptans (compounds of formula RSH) in a hydrocarbon containing stream.

[0082] The mercaptans which are eliminated are typically those of formula RSH wherein R is an alkyl or alkenyl group containing from 1 to 8, preferably from 1 to 6 and more preferably from 1 to 4 carbon atoms.

[0083] The hydrocarbon containing streams are typically selected from crude petroleum oils, hydrocarbon fractions and residues deriving from the distillation thereof, light petroleum gas (LPG) and natural gas, as well as mixtures thereof with aqueous phases such as brine.

[0084] Such streams contain H₂S and/or mercaptans in total amounts which may range for example from 1 to 10 000 ppm by weight.

[0085] The compound of the invention is used in a molar amount which depends on the total content of hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in the liquid or gaseous stream. Typically, the compound of the invention is used at a molar ratio of 0.2 to 5 moles per mole of hydrogen sulphide and of organic compounds comprising at least one sulfhydryl group in the liquid or gaseous stream, preferably 0.5 to 2 moles.

The method for scavenging hydrogen sulphide

[0086] The method of the invention consists in contacting a liquid or gaseous stream containing hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group with a compound as described above, or with a composition containing such as compound as described above.

[0087] As described above, the liquid or gaseous stream may be monophasic such as in particular an aqueous stream or an organic stream, or multiphasic such as a stream containing both water and hydrocarbons (such as for instance oil/water or oil/water/gas or gas/water).

[0088] According to a preferred embodiment, the stream contains hydrocarbons. The stream may be especially selected from crude petroleum oils, hydrocarbon fractions and residues deriving from the distillation thereof such as in particular fuel oils and heavy fuel oils, light petroleum gas (LPG) and natural gas, as well as mixtures thereof with aqueous compositions such as brine.

[0089] The amount of composition used per amount of stream depends on the concentration of scavenging active compound in the composition as well as the total content of hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in the liquid or gaseous stream, as explained above.

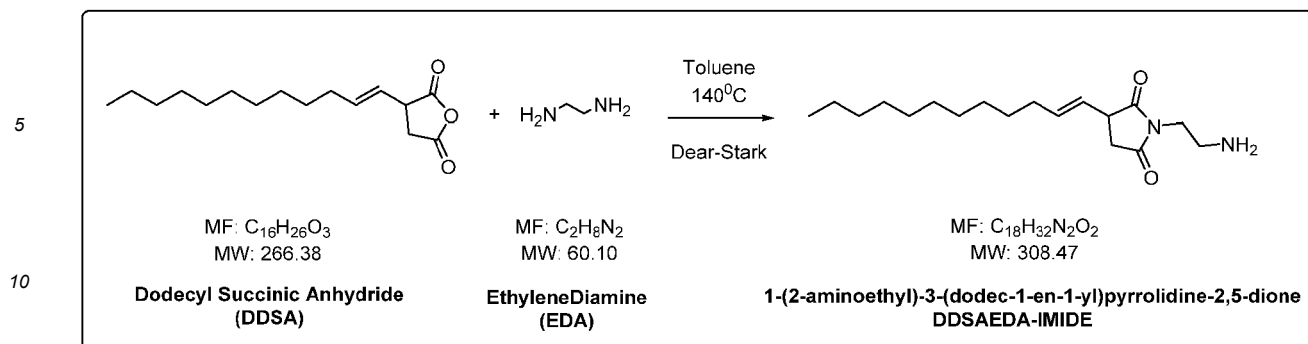
[0090] The composition is contacted with the liquid or gaseous stream for a time sufficient to achieve an effective scavenging of hydrogen sulphide and of organic compounds comprising at least one sulfhydryl group.

[0091] The example hereafter only aims at illustrating the present invention, and shall not be interpreted so as to limit its scope.

EXAMPLESExample 1: Synthesis of a first scavenging compound C1

[0092] Compound C1 according to the invention was prepared following the protocol detailed below.

[0093] First, a DDSAEDA-IMIDE compound (amino compound (i)) was prepared in accordance with the following reaction scheme and the synthesis procedure detailed hereunder:



15 **[0094]** Dodecyl succinic anhydride (DDSA) (40 g, 0.15 moles), was added dropwise over 1 hour to a solution of ethylene diamine (EDA) (10.8 g, 0.18 moles) in toluene (400 mL). The reaction mixture was heated to 140° C for 6 h and water released was collected and measured in Dean-Stark trap. The reaction product was isolated by evaporating all the solvent under reduced pressure. 48.0 g (99% yield) of desired product DDSAEDA-IMIDE was obtained as viscous brown liquid, in pure form, it was used as such for further steps without any purifications.

20 Spectral Data for the DDSAEDA-IMIDE compound:

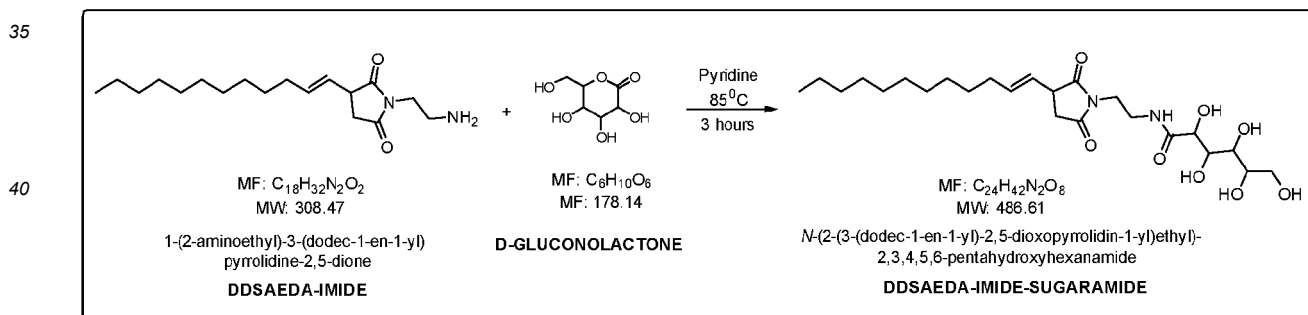
[0095]

25 FT-IR (cm⁻¹): 3455, 3305, 2956, 2930, 2871, 1772, 1693, 1667, 1545, 1432

¹H - NMR (300 MHz, DMSO-d₆): 8.0 (-NH₂, m, 2H); 4.9 - 4.6 (>C=CH₂, m, 2H); 3.3 (>N-CH₂-CH₂-NH₂), 3.5 (>N-CH₂-CH₂-NH₂), 2.7 (-CH₂-CH, m, 1H), 2.6 (-CH₂-CH, m, 2H); 1.9 (H₂C=C=, d, 2H); 1.6 - 1.2 (-CH₂, m, 16H); 1.0 - 0.6 (-CH₃, m, 3H).

¹³C - NMR (60 MHz, DMSO-d₆): 178.61, 176.40, 172.21, 167, C=O; 137.21, H₂C=CH₂; 49.45, 47.84, N-CH₂-CH₂-NH₂; 30.51, 29.99, 28.60, 22.84, 21.14, 18.56 alkyl carbons.

30 **[0096]** Secondly, a DDSAEDA-IMIDE-SUGARAMIDE compound was prepared in accordance with the following reaction scheme and the synthesis procedure detailed hereunder (step (a) of the method of the invention):



45 **[0097]** DDSAEDA-IMIDE (1.0 eq, 30.8 g, 0.1 mole) and gluconolactone (GL) (1.2 eq, 21.36 g, 0.12 moles) were added to pyridine (50 mL) and the reaction mixture was heated to 85° C for 3 h. The solution turned dark brown after 3 h. Pyridine from the resulting reaction mixture was removed on rotavapor under reduced pressure. The crude dark brown solid was subjected to water wash for 2 h followed by addition of toluene for water removal on rotavapor. On removal of water, product was obtained as a dark brown solid in an average 80 % yield. Resulted product was further purified by precipitation techniques to obtain DDSAEDA-IMIDE-SUGARAMIDE with more than 96% purity.

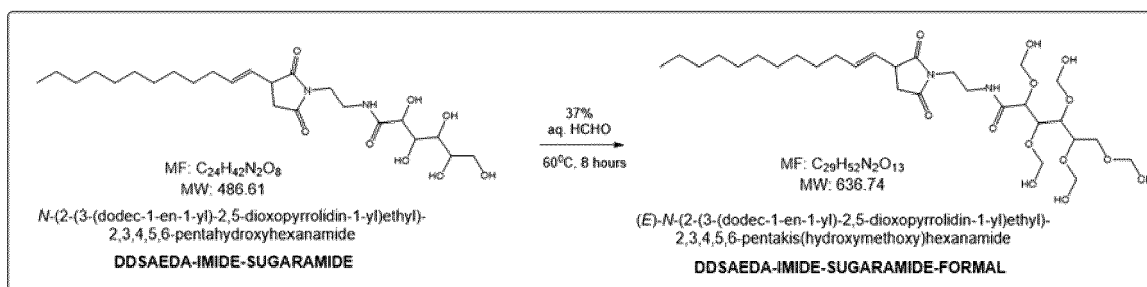
Spectral Data for the DDSAEDA-IMIDE-SUGARAMIDE compound:

55 **[0098]** FT-IR (cm⁻¹): 3349, 2956, 2930, 2871, 1769, 1693, 1649, 1544, 1433

[0099] Thirdly, scavenging compound C1 (DDSAEDA-IMIDE-SUGARAMIDE-FORMAL compound below) was obtained in accordance with the following reaction scheme and the synthesis procedure detailed hereunder (step (b) of the method of the invention):

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[0100] To a solution of 37% aq. formaldehyde was added DDSAEDA-IMIDE-SUGARAMIDE (12.72 g, 0.02 moles) followed by 2.0 g of 50% NaOH solution. The resultant solution was kept in glass vial for 24 h at 55°C in oven. Saturated brine was added to extract sodium hydroxide into aqueous phase and the organic phase was dried over sodium sulfate to obtain the product as a dark brown viscous liquid in an average 80 % yield. This product was characterized as DDSAEDA-IMIDE-SUGARAMIDE-FORMAL.

Spectral Data for the DDSAEDA-IMIDE-SUGARAMIDE-FORMAL compound C1:

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[0101] FT-IR (cm⁻¹): 3374, 2958, 2872, 2108, 1633, 1560, 1459, 1402

Example 2: Synthesis of a second scavenging compound C2

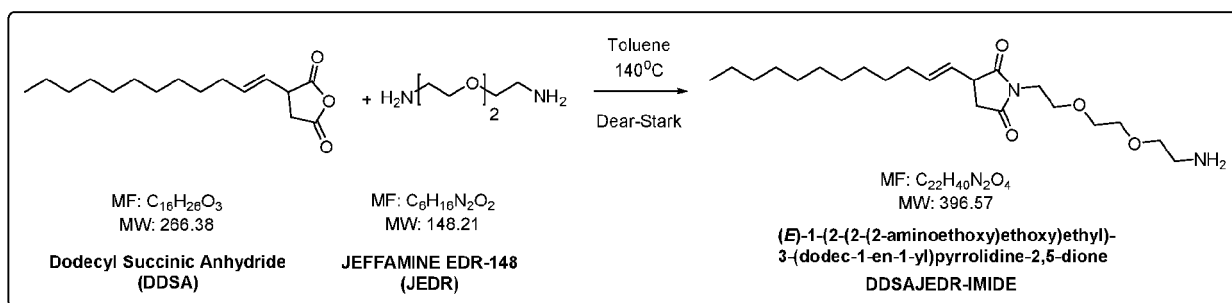
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[0102] Compound C2 according to the invention was prepared following the protocol detailed below.

[0103] First, a DDSAJEDR-IMIDE compound (amino compound (i)) was prepared in accordance with the following reaction scheme and the synthesis procedure detailed hereunder:

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[0104] Dodecyl succinic anhydride (DDSA) (40 g, 0.15 moles), was added dropwise over 1 hour to a solution of Jeffamine-148 (JEDR) (26.64 g, 0.18 moles) in toluene (400 mL). The reaction mixture was heated to 140° C for 6 h and water released was collected and measured in Dean-Stark trap. The reaction product was isolated by evaporating all the solvent under reduced pressure. 56.0 gm (99% yield) of desired product DDSAJEDR-IMIDE was obtained as viscous brown liquid, in pure form, it was used as such for further steps without any purifications.

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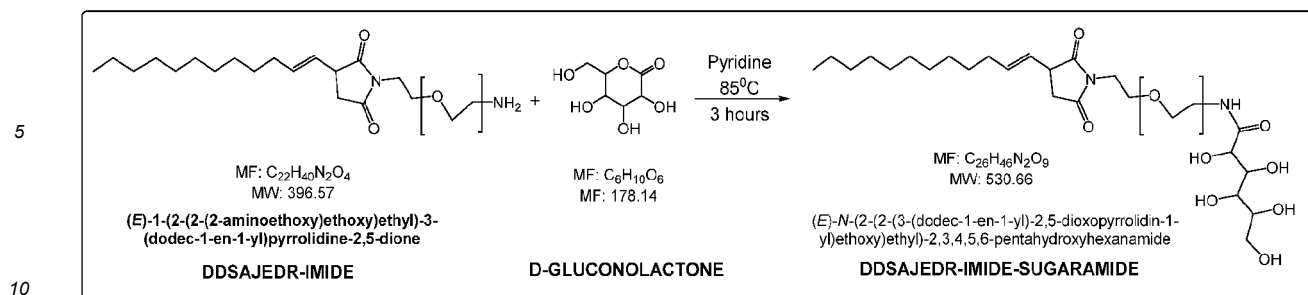
Spectral Data for the DDSAJEDR-IMIDE compound:

[0105] FT-IR (cm⁻¹): 2956, 2869, 1772, 1697, 1545, 1434, 1397, 1330

[0106] Secondly, a DDSAJEDR-IMIDE-SUGARAMIDE compound was prepared in accordance with the following reaction scheme and the synthesis procedure detailed hereunder (step (a) of the method of the invention):

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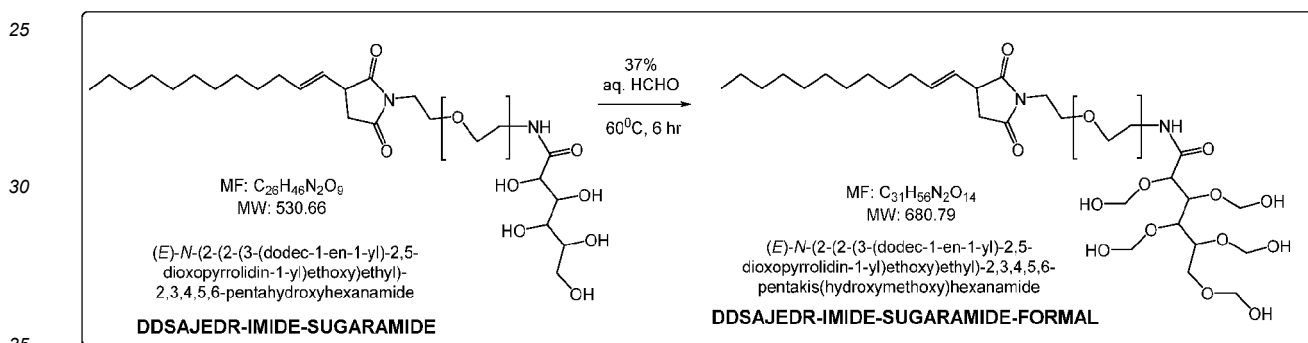
[0107] The DDSAJEDR-IMIDE-SUGARAMIDE compound was synthesized following exactly the same procedure as described above for the DDSAEDA-IMIDE-SUGARAMIDE compound of Example 1.

15 [0108] 39.6 g (0.1 mole) of DDSAJEDR-IMIDE was reacted with 18.8 g (0.1 mole) of D-gluconolactone in pyridine, which resulted, 45 g of DDSAJEDR-IMIDE-SUGARAMIDE as dark brown liquid.

Spectral Data for the DDSAJEDR-IMIDE-SUGARAMIDE compound:

20 [0109] FT-IR (cm⁻¹): 3363, 2957, 2873, 1695, 1640, 1548, 1401, 1332, 1193, 1089

[0110] Thirdly, scavenging compound C2 (DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL compound below) was obtained in accordance with the following reaction scheme and the synthesis procedure detailed hereunder (step (b) of the method of the invention):



[0111] The DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL compound was synthesized following exactly the same procedure as described above for the DDSAEDA-IMIDE-SUGARAMIDE-FORMAL compound of Example 1.

40 [0112] 26.5 g (0.05 moles) of DDSAJEDR-IMIDE-SUGARAMIDE was reacted with 25 g (0.3 moles) of 37% aq. formaldehyde solution in presence of 50% aq. NaOH solution. DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL was obtained as heavy viscous dark brown material.

Spectral Data for the DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL compound C2:

45 [0113] FT-IR (cm⁻¹): 3381, 2957, 2930, 2872, 2123, 1636, 1556, 1458, 1400

Example 3: Assessment of H₂S scavenging ability of compounds C1 and C2

50 [0114] The test was carried out at room temperature. A 250 mL glass purger was charged with 100 mL of an inert solvent. A gas consisting of N₂ containing H₂S at a concentration of 50 ppm was released by a bubbling tube at a flow rate of 0.4 mL/min into the solvent contained in said purger. The solvent was continuously agitated by using a magnetic stirrer. The gas exiting the solvent and leaving the purger was conveyed to a H₂S detector.

55 [0115] At the beginning of each test, a setup was made as follows: the gas (H₂S in N₂ at a concentration of 50 ppm) was passed through the purger with a 0.4 mL/min flow rate, and the H₂S detector started recording the H₂S concentration at the outlet of the purger. Once the outlet H₂S concentration remained stable (with a value of 50 ppm constant reading at the detector), a solution consisting of 0.5g of tested scavenging compound C in 0.5 g of butyl carbitol was quickly injected into the purger. As scavenging compound C dissolves within the solvent, it starts reacting with H₂S which results in a reduction of the H₂S concentration at the outlet, as recorded by the detector. The H₂S concentration was recorded

at every 5 sec time interval.

[0116] An interval of time of 30 min was respected between each test. In each test, once the H₂S scavenger has been injected into the purger, the concentration of H₂S recorded by the detector started decreasing from 50 ppm down to potentially 0 ppm, depending on the efficiency of the scavenging compound C. The higher the H₂S scavenging efficiency of compound C, the faster the H₂S concentration recorded by the detector drops from 50 ppm to 0 ppm and the longer the rate of H₂S concentration recorded by the detector remains at 0 ppm. It was also observed that, once the scavenging compound C was consumed, the concentration of H₂S recorded by the detector started increasing slowly back to 50 ppm.

[0117] The following compounds C were tested:

- Comparative, prior art: 3,3'-methylenebis(5-methyloxazolidine) also known as MBO;
- Comparative: DDSAEDA-IMIDE-SUGARAMIDE as prepared in example 1;
- Comparative: DDSAJEDR-IMIDE-SUGARAMIDE as prepared in example 2;
- Inventive compound C1: DDSAEDA-IMIDE-SUGARAMIDE-FORMAL as prepared in example 1;
- Inventive compound C2: DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL as prepared in example 2.

[0118] The results obtained are detailed in the Table below:

Tested compound C	Min H ₂ S recorded	% H ₂ S scavenging efficiency
MBO	16 ppm	68 %
DDSAEDA-IMIDE-SUGARAMIDE	47 ppm	6 %
DDSAJEDR-IMIDE-SUGARAMIDE	46 ppm	8 %
DDSAEDA-IMIDE-SUGARAMIDE-FORMAL C1	0 ppm	100 %
DDSAJEDR-IMIDE-SUGARAMIDE-FORMAL C2	0 ppm	100 %

[0119] The above results show that inventive compounds C1 and C2 exhibit an excellent efficiency in scavenging H₂S. The performances achieved are much above those obtained with the comparative compounds, including MBO which a well known hydrogen sulphide scavenger.

Claims

1. A compound obtainable by reaction of at least:

- (i) an amino compound comprising at least one primary amine group, this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with
- (ii) a polyhydroxy carboxylic acid or an ester thereof; and then with
- (iii) formaldehyde.

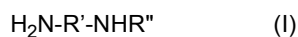
2. The compound as defined in the preceding claim, wherein said acylating agent is selected from mono- or poly-carboxylic acids and their ester, amide or anhydride derivatives, preferably from succinic, phthalic and propionic acids and the corresponding anhydrides, and more preferably the acylating agent is succinic anhydride.

3. The compound as defined in any preceding claim, wherein said hydrocarbon group containing at least 8 carbon atoms is chosen from:

- linear or branched, saturated or unsaturated aliphatic C₈-C₂₂ hydrocarbon groups, more preferably from C₈-C₂₂ alkyl or mono or polyunsaturated alkenyl groups; and
- homo- and co-polymers of mono- and di-olefins having from 2 to 10 carbon atoms, preferably from polyisobutenyl groups (PIB groups) having preferably, a number average molecular weight (Mn) between 170 and 2800, preferably between 250 and 1500, even more preferably between 500 and 1500.

4. The compound as defined in any preceding claim, wherein said compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols is chosen from the

following amines of formula (I) and (II):



5 and



10 wherein

R^I denotes a C₁ to C₁₀, preferably C₁ to C₄ hydrocarbon chain,
 R^{II} denotes hydrogen or a C₁ to C₄ alkyl group,
 and preferably from diamines of formula (I), and more preferably those wherein R^{II} denotes hydrogen.

- 15 **5.** The compound as defined in any preceding claim, wherein said polyhydroxy carboxylic acid (ii) is chosen from those containing from 3 to 10 carbon atoms, more preferably from 4 to 8 carbon atoms, and bearing at least two hydroxy groups.
- 20 **6.** The compound as defined in any preceding claim, wherein said compound (ii) is chosen from sugar acids and esters thereof, including internal esters (lactone forms), more preferably from aldonic acids such as glyceric acid, xylonic acid, gluconic acid, and gluconolactone, and more preferably said compound (ii) is δ-gluconolactone.
- 25 **7.** The compound as defined in any preceding claim, obtainable by further reacting the condensation product of compounds (i), (ii) and (iii) with (iv) a quaternizing agent, which is preferably selected from dialkyl sulfates, carboxylic acid esters, alkyl halides, benzyl halides, hydrocarbon carbonates, and hydrocarbon epoxides optionally mixed with an acid.
- 30 **8.** A composition comprising at least one liquid solvent and at least one compound as defined in any one of the preceding claims.
- 35 **9.** The composition as defined in the preceding claim, wherein the solvent is chosen from water and organic solvents, preferably from aromatic solvents, poly alkyl ethers and organic solvents derived from biomass, more preferably from oils of vegetable origin, and even more preferably from cashew nutshell liquid and mixtures thereof with other organic solvents.
- 10.** The composition as defined in any one of claims 8 and 9, containing said at least one compound obtained by reaction of (i), (ii), (iii) and optionally (iv) in an amount ranging from 5 to 75% by weight, preferably from 10 to 60% by weight, more preferably from 20 to 50% by weight, relative to the total weight of the composition.
- 40 **11.** Use of a compound as defined in any one of claims 1 to 7 for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream.
- 12.** Use as defined in claim 11, for scavenging hydrogen sulphide and mercaptans in a hydrocarbon containing stream.
- 45 **13.** Use as defined in any one of claims 11 and 12, wherein said liquid or gaseous stream is chosen from hydrocarbon containing streams, more preferably from crude petroleum oils, hydrocarbon fractions and residues deriving from the distillation thereof, light petroleum gas and natural gas, as well as mixtures thereof with aqueous phases.
- 50 **14.** Use as defined in any one of claims 11 to 13, wherein said compound is used in a molar ratio of 0.2 to 5 mole(s) per mole of hydrogen sulphide and of organic compounds comprising at least one sulfhydryl group in the liquid or gaseous stream, preferably 0.5 to 2 mole(s) per mole of hydrogen sulphide and of organic compounds comprising at least one sulfhydryl group in the liquid or gaseous stream.
- 55 **15.** A method for scavenging hydrogen sulphide and/or organic compounds comprising at least one sulfhydryl group in a liquid or gaseous stream containing them, comprising contacting said stream with a compound as defined in any one of claims 1 to 7, or with a composition as defined in any one of claims 8 to 10.
- 16.** A method of preparation of a compound, comprising the following steps:

EP 4 279 566 A1

(a) a step of condensation of an amino compound (i) comprising at least one primary amine group, this amino compound being the product of the reaction of an acylating agent substituted by a hydrocarbon group containing at least 8 carbon atoms and of a compound comprising at least one primary amine group and at least one group selected from primary amines, secondary amines and alcohols; with a polyhydroxy carboxylic acid or an ester thereof (ii) ; followed by

(b) a step of condensation of the product of step (a) with formaldehyde (iii); optionally followed by
(c) a step of quaternization of the compound resulting from step (b), by reacting the product of step (b) with a quaternizing agent (iv) which is preferably selected from dialkyl sulfates, carboxylic acid esters, alkyl halides, benzyl halides, hydrocarbon carbonates, and hydrocarbon epoxides optionally mixed with an acid.

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EUROPEAN SEARCH REPORT

Application Number
EP 22 30 5749

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DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
A	<p>WO 2020/115132 A1 (TOTAL MARKETING SERVICES [FR]) 11 June 2020 (2020-06-11) * claim 1 *</p> <p style="text-align: center;">-----</p>	1-16	<p>INV. C10G29/20 C10L3/10</p>
			<p>TECHNICAL FIELDS SEARCHED (IPC)</p> <p>C10G C10L</p>
<p>The present search report has been drawn up for all claims</p>			
<p>Place of search</p> <p>Munich</p>		<p>Date of completion of the search</p> <p>5 October 2022</p>	<p>Examiner</p> <p>Klaes, Daphne</p>
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document</p>			

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**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 22 30 5749

5 This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

05-10-2022

10	Patent document cited in search report	Publication date	Patent family member(s)	Publication date
15	WO 2020115132 A1	11-06-2020	EP 3891260 A1	13-10-2021
			US 2022041944 A1	10-02-2022
			WO 2020115132 A1	11-06-2020
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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- WO 9802501 A [0008]
- WO 2017102693 A [0010]
- EP 0565285 A [0041]
- EP 1344785 A [0041]