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**USE OF A FATTY AMINE FOR PREVENTING AND/OR REDUCING THE METAL
LOSSES OF THE PARTS IN AN ENGINE**

The present invention relates to the prevention and/or the reduction of metal losses of the parts of an engine, in particular a marine engine. More particularly, the present invention relates to the prevention and/or the reduction of metallic losses of parts of a marine engine due to the contact of said parts with an acid medium.

The subject matter of the present invention is the use of one or a plurality of soluble fatty amines in a lubricant composition to prevent and/or reduce metallic losses of parts of an engine, in particular a marine engine.

The subject matter of the present invention is also a method for preventing and/or reducing metallic losses of parts of an engine, in particular a marine engine wherein said parts are put in contact with one or a plurality of soluble fatty amines in a lubricant composition.

The combustion of fuel oils generates the formation of acid gases, in particular sulfur oxides (SO_2 , SO_3). Said acid gases make amongst others, fuel oil combustion residues; said residues are in contact with the lubricant oil and consequently are also in contact with parts of the engine. When in contact with moisture present in the combustion gases and/or in the lubricant oil, said acid gases hydrolyze into sulfurous acid (HSO_3) or sulfuric acid (H_2SO_4) that in turn are in contact with parts of the engine.

In the case of marine engines, in particular two-stroke marine engines, lubricant oils are classified into two categories: on the one hand, cylinder oils, that ensure the lubrication of the piston-cylinder assembly, and on the other hand, system oils that ensure the lubrication of all moving parts other than the same of the piston-cylinder assembly. More precisely, it is within the cylinder piston assembly that the combustion residues containing acid gases are in contact with the lubricant oil.

In general, the neutralization of said acids occurs by reaction with basic sites included in the lubricant. The neutralization ability of an oil is measured by the BN or Base Number thereof, characterizing the basicity thereof. The Base Number is measured according to the standard ASTM D- 2896 and is expressed as the equivalent by weight of potash per gram of oil or mg of KOH/g of oil. The BN is a classic criterion allowing the basicity of cylinder oils to be adjusted to the sulfur concentration of the fuel oil used, in order to be able to neutralize all the sulfur contained in the fuel, and likely to be transformed into sulfuric acid by combustion and hydrolysis.

More precisely, marine oils available on the market have a BN varying from 5 to 130 mg KOH/g of oil. This basicity is in particular furnished by detergents which are overbased by insoluble metallic salts, in particular metallic carbonates. The detergents, mainly of the anionic type, are e.g. metallic soaps of the salicylate, phenate, sulfonate, carboxylate, etc. type, which form micelles where insoluble metallic salts are maintained in suspension. The usual overbased detergents intrinsically have a BN traditionally comprised between 150 and 700 mg of potash per gram of detergent.

A portion of the BN can also be furnished by non-overbased or "neutral" detergents with BN typically less than 150 mg of potash per gram of detergent.

Nevertheless, the claimant has observed that during the combustion of fuel oil, neutral and/or overbased detergents present in the lubricant composition deteriorate chemically and consequently form ashes, also called residues or deposits that favor the clogging of the engine, in particular a marine engine.

In order to decrease the amount of ashes formed during the combustion of fuel oil, the claimant replaced a portion of the detergents furnishing all the BN of the lubricant composition by compounds furnishing the BN and not forming or forming few ashes during the combustion of fuel oil. The claimant therefore has developed lubricant compositions wherein a portion of detergents furnishing the BN of the lubricant composition is replaced by amine containing compounds.

WO 2009/153453 describes a lubricant composition for a cylinder having a BN greater than or equal to 40 milligrams of potash per gram of lubricant and comprising a base oil, an alkali or alkaline earth metal based detergent, overbased by metal carbonate salts, a neutral detergent and a fatty amine and/or an oil soluble fatty amine derivative having a BN comprised between 150 and 600 milligrams of potash per gram of lubricant oil.

WO 2014/180843 describes a lubricant composition for a cylinder having a BN greater than or equal to 50 milligrams of potash per gram of lubricant and comprising a base oil, an alkali or alkaline earth metal based detergent, overbased by metal carbonate salts, a neutral detergent and a mixture of fatty amines having four amine groups.

EP 2 486 113 describes a lubricant oil for a marine engine comprising a monoamine comprising a base oil, a detergent and a monoamine, di-substituted by a hydrogen atom or a hydrocarbon moiety having from 1 to 50 carbon atoms.

WO 2011/042552 describes a lubricant oil for engines comprising a base oil, a detergent and a monoamine, di-substituted by a hydrocarbon group comprising from 1 to

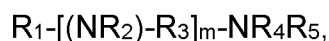
50 carbon atoms and by a hydrogen atom or a hydrocarbon moiety comprising from 1 to 50 atoms of carbon.

In the continuity of the Claimant's research, the Claimant discovered surprisingly that the amine compounds, usually used for replacing a portion of the BN of the lubricant composition while decreasing the amount of ash formed during the combustion of fuel oil, also allows metallic losses of parts of the engine, in particular of a marine engine, to be prevented and/or reduced, when said parts are in contact with acids coming from the combustion of fuel oil.

Hence, the claimant company has noted that the use of one or a plurality of soluble fatty amines in a lubricant composition,

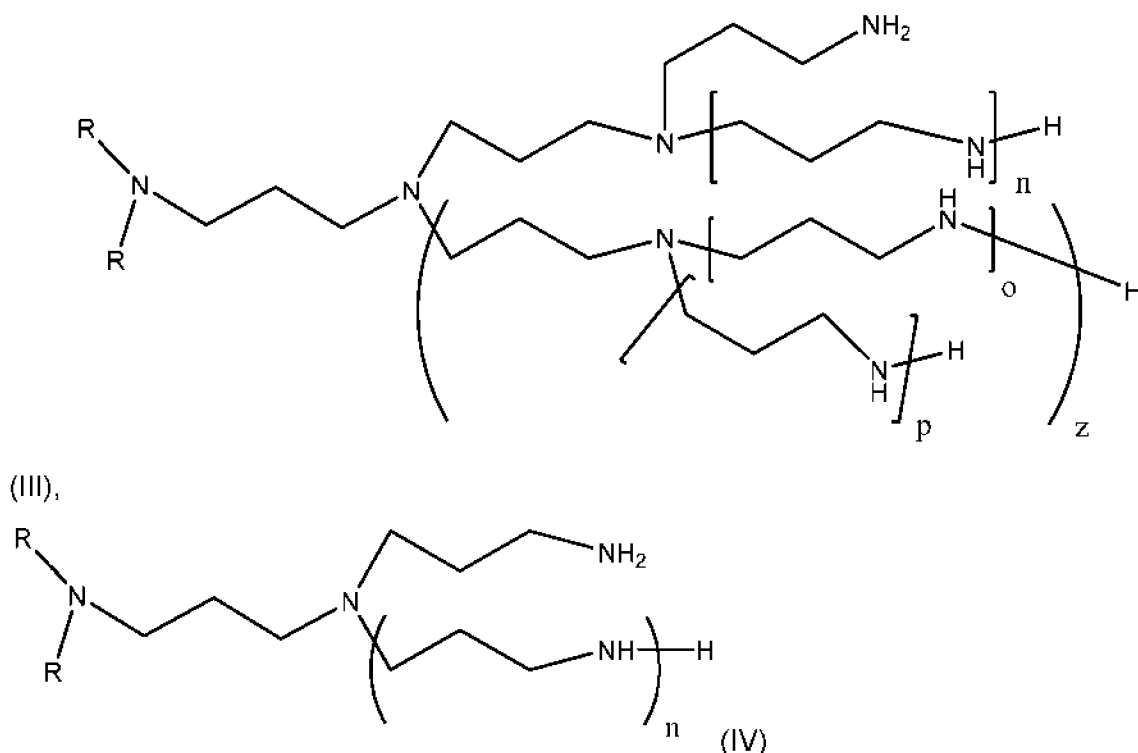
the fatty amine being chosen from:

- compounds with the formula (I):



wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently, a hydrogen atom; a saturated or unsaturated, linear or branched, alkyl moiety comprising between 1 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,
- R_3 represents a saturated or unsaturated, linear or branched, alkyl moiety comprising between 2 and 6 carbon atoms,
- m is a whole number comprised between 1 and 10, more preferably between 1 and 6, even more preferably chosen from 1, 2 or 3, or
- a mixture of fatty polyalkylamines comprising one or a plurality of polyalkylamines with the formulae (III) and/or (IV):



wherein

- R, identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z independently of each other, represent 0, 1, 2 or 3 and
- when z is greater than 0, o and p represent independently of each other, 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or the derivatives thereof, or

- the mixtures of fatty amines of formulae (I), (III) and/or (IV)

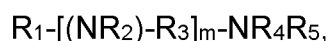
allow metallic losses of parts of an engine, preferably a marine engine, to be prevented and/or reduced.

Fatty amines of formula (I) comprised in a lubricant composition are known as such in the applications WO 2009/153453 and WO 2014/180843 filed by the claimant. The claimant has now discovered a new use for said fatty amines.

The present application describes the use of one or a plurality of soluble fatty amines in a lubricant composition for preventing and/or reducing metallic losses from metal parts of an engine, preferably a marine engine,

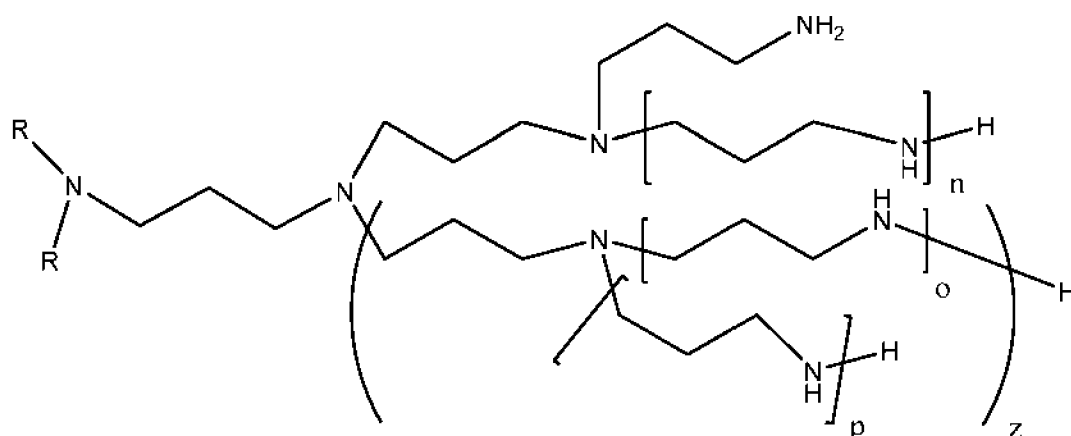
the fatty amine being chosen from:

- compounds with the formula (I):

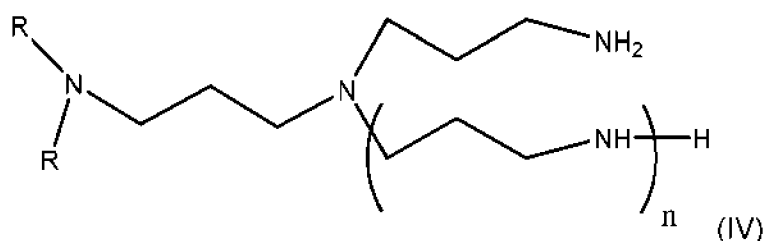


wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising at least 12 carbon atoms, and optionally at least one heteroatom chosen from nitrogen, sulfur or oxygen,
 - R_2 , R_4 or R_5 represent independently, a hydrogen atom or a saturated or unsaturated, linear or branched hydrocarbon moiety and optionally comprising at least one heteroatom chosen from nitrogen, sulfur or oxygen,
 - R_3 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising one or a plurality of carbon atoms, and optionally comprising at least one heteroatom chosen from nitrogen, sulfur or oxygen, preferably oxygen,
 - m is a whole number greater than or equal to 1, preferably comprised between 1 and 10, more preferably between 1 and 6, even more preferably being chosen from 1, 2 or 3; or
- a mixture of fatty di-alkyl polyalkylamines comprising one or a plurality of polyalkylamines of formula (III) and/or (IV):



(III),



wherein

- R , identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z independently of each other, represent 0, 1, 2 or 3, and

- when z is greater than 0, o and p represent independently of each other, 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or of their derivatives; or

- a mixture of fatty amines of formulae (I), (III) and/or (IV).

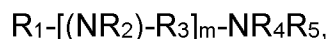
Without being related to the theory, the claimant has observed surprisingly that the fatty amines according to the invention allow not only to furnish organic BN to the lubricant composition while furnishing little or no ashes during the combustion of fuel oil but also allowing to prevent and/or reduce metallic losses of parts of an engine, in particular a marine engine, when said parts are put in contact with acids coming from the combustion of fuel oil. More precisely, the claimant has discovered unexpectedly that said soluble fatty amines in a lubricant composition in the presence of a large excess of sulfuric acid allow metallic losses of parts directly in contact with said lubricant composition and the large excess of sulfuric acid to be limited and/or reduced. This limitation and/or reduction of metallic losses of parts is probably due to the passivation of all or part of the surface of the metallic parts by said fatty amines.

Within the meaning of the invention, "metallic losses of parts of an engine" means metallic losses coming from the attack of said parts by acids and not metallic losses generated by friction of one metallic part on another.

The present invention relates to the use of one or a plurality of soluble fatty amines in a lubricant composition for passivating all or part of the surface of metallic parts of an engine, preferably a marine engine, and preventing and/or reducing the metallic losses from said metallic parts,

the fatty amine is chosen from:

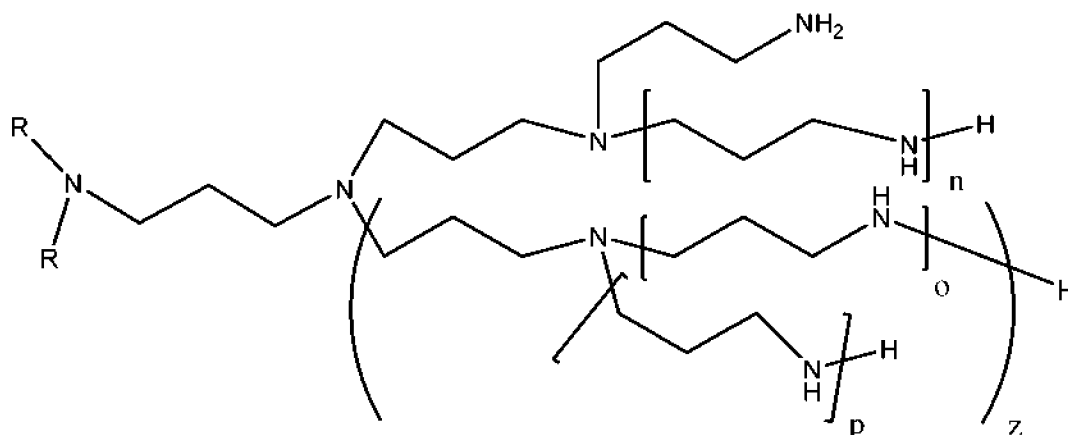
- The compounds of formula (I):



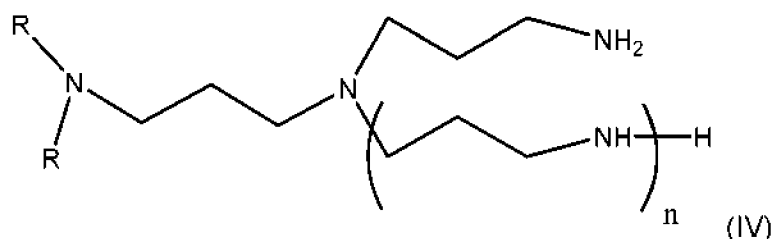
wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently, a hydrogen atom; a saturated or unsaturated, linear or branched, alkyl moiety comprising between 1 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated or unsaturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,

- R_3 represents a saturated or unsaturated, linear or branched, alkyl moiety comprising between 2 and 6 carbon atoms;
- m is a whole number comprised between 1 and 10, more preferably between 1 and 6, even more preferably is chosen from 1, 2 or 3; or
- a mixture of one or a plurality of polyalkylamines with the formulae (III) and/or (IV):



(III),



wherein

- R , identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z independently of each other, represent 0, 1, 2 or 3 and
- when z is greater than 0, o and p independently of each other, represent 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or of their derivatives; or

- a mixture of fatty amines of formulae (I), (III) and/or (IV).

Preferably, when the fatty amine is of the formula (I):

- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 12 to 22 carbon atoms, preferably from 14 to 22 carbon atoms and optionally at least one heteroatom chosen from nitrogen, sulfur or oxygen, and/or

- R_2 , R_4 or R_5 represent independently, a hydrogen atom; a saturated or unsaturated, linear or branched alkyl moiety comprising between 1 and 22 carbon atoms, preferably between 14 and 22 carbon atoms, more preferably between 16 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety, comprising at least 2 carbon atoms, preferably between 2 and 6 carbon atoms, more preferably between 2 and 4 carbon atoms and q represents a whole number greater than or equal to 1, preferably comprised between 1 and 6, more preferably comprised between 1 and 4, and/or
- R_3 represents a saturated or unsaturated, linear or branched, alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms.

More preferably when the fatty amine is of formula (I):

- m is equal to 1, 2 or 3,
- R_1 represents a saturated or unsaturated, either linear or branched alkyl moiety comprising from 12 to 20 carbon atoms, preferably from 14 to 20 carbon atoms and optionally at least one heteroatom chosen from nitrogen, sulfur or oxygen,
- R_2 represents independently a hydrogen atom or a saturated, linear or branched alkyl moiety comprising from 1 to 20 carbon atoms, preferably from 16 to 20 carbon atoms, more preferably from 16 to 18 carbon atoms,
- R_3 represents a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms.
- R_4 and R_5 represent a hydrogen atom or a methyl moiety, preferentially a hydrogen atom.

Advantageously, when the fatty amine is of formula (I):

- m is equal to 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 12 to 20 carbon atoms, preferably from 14 to 20 carbon atoms, more preferably from 16 to 20 carbon atoms and optionally at least one heteroatom chosen from nitrogen, sulfur or oxygen,
- R_2 represents independently a hydrogen atom or a saturated, linear or branched alkyl moiety, comprising from 16 to 18 carbon atoms,
- R_3 represents an ethyl or propyl moiety,

- R_4 and R_5 represent a hydrogen atom.

More preferably when the fatty amine is also of formula (I):

- m is equal to 1, 2 or 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 , R_4 and R_5 represent independently a hydrogen atom, or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, more preferably between 2 and 4 carbon atoms and q representing a whole number comprised between 1 and 6, more preferably comprised between 1 and 4.
- R_3 represents a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms.

Advantageously, when the fatty amine is also of formula (I):

- m is equal to 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 , R_4 and R_5 represent independently a hydrogen atom, or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear alkyl moiety comprising between 2 and 4 carbon atoms, and q representing a whole number comprised between 1 and 4.
- R_3 represents an ethyl or propyl moiety.

According to a particular embodiment of the invention, the use of a fatty amine of formulae (I), (III) and/or (IV) allows metallic losses of parts of a two-stroke or four-stroke marine engine to be prevented and/or reduced during the combustion of fuel oil.

Preferably, the use of a fatty amine of formulae (I), (III) and/or (IV) allows metallic losses of parts in the hot parts, in particular the piston-jacket segment zone of a two-stroke or four-stroke, marine engine to be prevented and/or reduced during the combustion of fuel oil.

Preferably, the fuel oil has a sulfur concentration less than 3.5% by weight with respect to the total weight of the fuel oil.

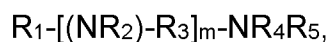
Detailed description of the invention.

Fatty amine

The subject matter of the invention relates to the use of one or a plurality of soluble fatty amines in a lubricant composition for preventing and/or reducing metallic losses from metal parts of an engine, preferably a marine engine,

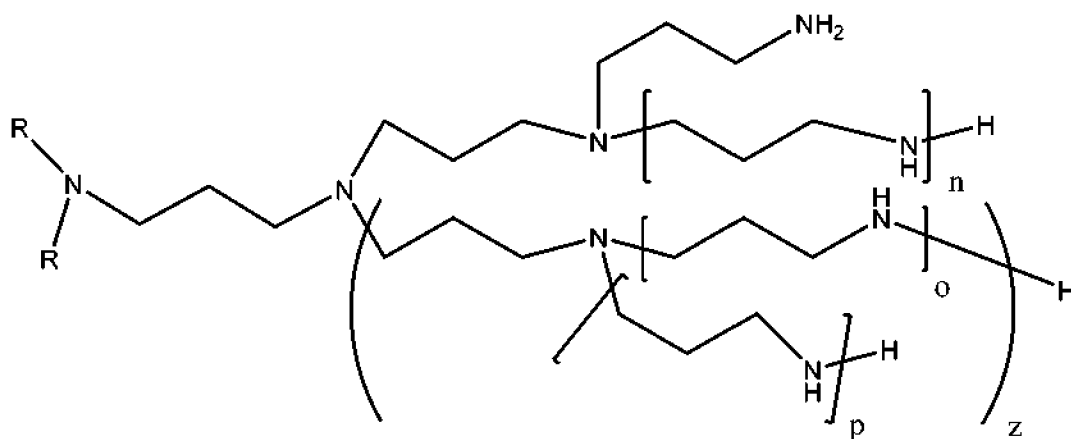
the fatty amine is chosen from:

- compounds with the formula (I):

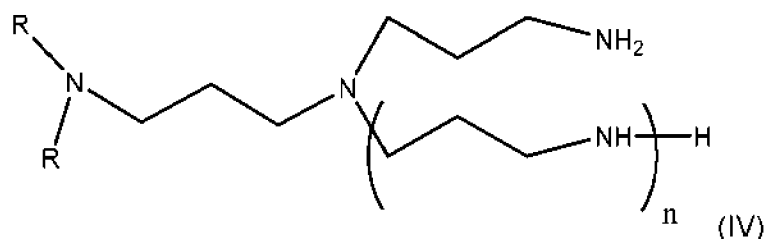


wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently a hydrogen atom, or a saturated or unsaturated, linear or branched alkyl moiety, comprising between 1 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,
- R_3 represents a saturated or unsaturated, linear or branched, alkyl moiety comprising between 2 and 6 carbon atoms,
- m is a whole number comprised between 1 and 10, more preferably between 1 and 6, even more preferably being chosen from 1, 2 or 3; or
- a mixture of fatty di-alkyl polyalkylamines comprising one or a plurality of polyalkylamines of formula (III) and/or (IV):



(III),



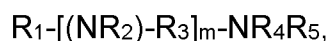
wherein

- R, identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z represent, independently of each other, 0, 1, 2 or 3, and
- when z is greater than 0, o and p represent independently of each other, 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or of their derivatives; or

- a mixture of fatty amines of formulae (I), (III) and/or (IV).

Preferably, another subject matter of the invention relates to the use of one or a plurality of soluble fatty amines in a lubricant composition for preventing and/or reducing metallic losses of parts of an engine, preferably a marine engine, the fatty amine being of formula (I):



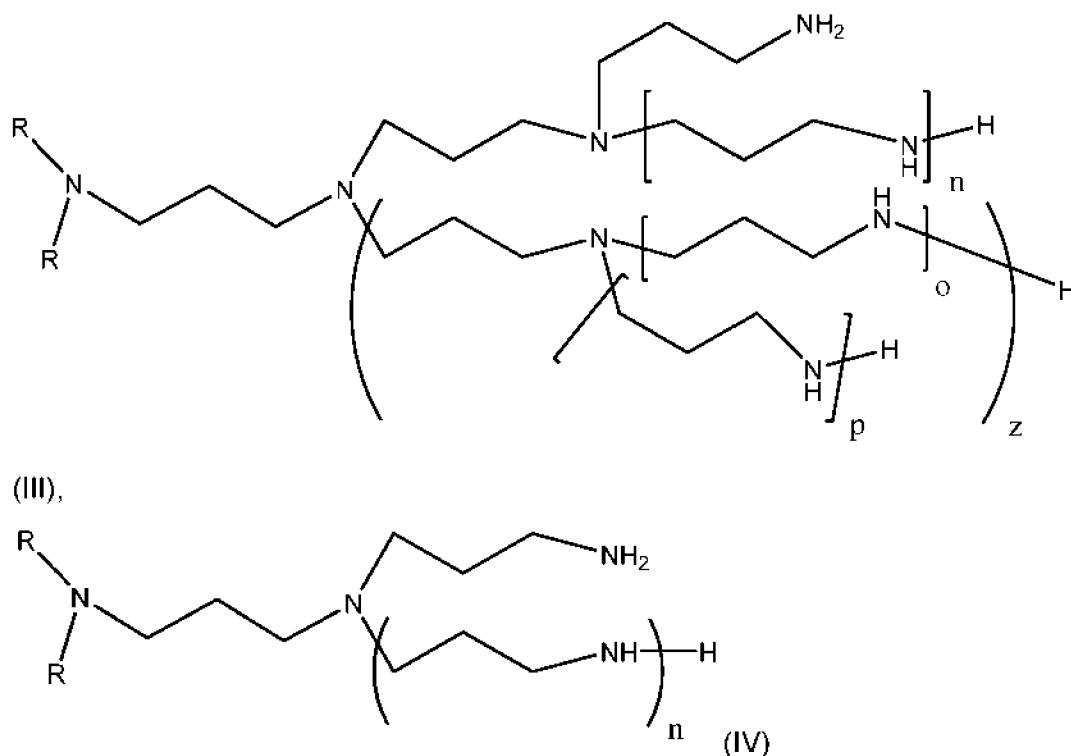
wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently, a hydrogen atom; a saturated or unsaturated, linear or branched, alkyl moiety comprising between 1 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,
- R_3 represents a saturated or unsaturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms,

m is a whole number comprised between 1 and 10, more preferably between 1 and 6, even more preferably being chosen from 1, 2 or 3.

Preferably, another subject matter of the invention relates to the use of one or a plurality of soluble fatty amines in a lubricant composition for preventing and/or reducing metallic losses of parts of an engine, preferably a marine engine, the fatty amine being a

mixture of fatty polyalkylamines comprising one or a plurality of polyalkylamines with the formulae (III) and/or (IV):



wherein

- R, identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z represent, independently of each other, 0, 1, 2 or 3, and
- when z is greater than 0, o and p represent independently of each other, 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or the derivatives thereof.

By “fatty amine” according to the invention is understood an amine of formula (I), (III) or (IV) comprising one or a plurality of saturated or unsaturated, linear or branched hydrocarbon moieties and optionally comprising at least one heteroatom chosen from amongst nitrogen, sulfur or oxygen, preferably oxygen.

By “a plurality of fatty amines” according to the invention is understood a mixture of fatty amines including at least one fatty amine of formula (I), (III) and/or (IV).

Preferably when the fatty amine is of formula (I):

- R₁ represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 12 to 22 carbon atoms, preferably from 14 to 22 carbon atoms, and/or

- R_2 , R_4 or R_5 represent independently, a hydrogen atom; a saturated or unsaturated, linear or branched alkyl moiety comprising between 1 and 22 carbon atoms, preferably between 14 and 22 carbon atoms, more preferably between 16 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety, comprising between 2 and 4 carbon atoms, and q represents a whole number greater than or equal to 1, preferably comprised between 1 and 6, more preferably comprised between 1 and 4, and/or
- R_3 represents a saturated or unsaturated, linear or branched, alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms.

Advantageously, when the fatty amine is of formula (I):

- m is equal to 1, 2 or 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 represents independently a hydrogen atom, linear or branched alkyl moiety comprising from 1 to 20 carbon atoms, more preferably from 16 to 20 carbon atoms, more preferably from 16 to 18 carbon atoms,
- R_3 represents a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms,
- R_4 and R_5 represent a hydrogen atom or a methyl moiety, preferentially a hydrogen atom.

In particular, when the fatty amine is of formula (I):

- m is equal to 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 represents independently a hydrogen atom or a saturated, linear or branched alkyl moiety, comprising from 16 to 18 carbon atoms,
- R_3 represents an ethyl or propyl moiety,
- R_4 and R_5 represent a hydrogen atom.

Advantageously, when the fatty amine is of formula (I):

- m is equal to 1, 2 or 3,

- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 , R_4 and R_5 represent independently a hydrogen atom, or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, more preferably between 2 and 4 carbon atoms and q representing a whole number comprised between 1 and 6, more preferably comprised between 1 and 4,
- R_3 represents a saturated, linear alkyl moiety comprising between 2 and 6 carbon atoms, preferably between 2 and 4 carbon atoms.

In particular, when the fatty amine is of formula (I):

- m is equal to 3,
- R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms,
- R_2 , R_4 and R_5 represent independently a hydrogen atom, or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear alkyl moiety comprising between 2 and 4 carbon atoms, and q representing a whole number comprised between 1 and 4,
- R_3 represents an ethyl or propyl moiety.

In general, fatty amines of formula (I) according to the invention are mainly obtained from carboxylic acids. These acids are dehydrated in the presence of ammonia for yielding nitriles and then undergo a catalytic hydrogenation in particular for leading to fatty amines.

As defined by the invention the fatty amine of formula (I) is obtained from at least one carboxylic acid, preferably from at least a fatty acid.

As defined by the invention, the alkyl group of the fatty amine of formula (I) has a number of carbon atoms corresponding to the number of carbon atoms of the carbon chain of the carboxylic acid, preferably corresponding to the number of carbon atoms of the carbon chain of the fatty acid.

As defined by the invention, a same fatty amine of formula (I) can be substituted by a plurality of alkyl moieties obtained from a plurality of carboxylic acids, identical or different, preferably obtained from a plurality of fatty acids, identical or different.

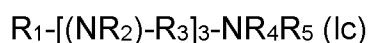
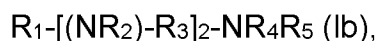
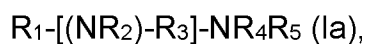
According to a particular embodiment of the invention, the alkyl moiety is obtained from the fatty acid chosen amongst the caprylic, pelargonic, capric, undecylenic, lauric,

tridecylenic, myristic, pentadecylic, palmitic, margaric, stearic, nonadecylic, arachic, heneicosanoic, behenic, tricosanoic, lignoceric, pentacosanoic, cerotic, heptacosanoic, montanic, nonacosanoic, melissic, hentriacontanoic, lacceroic acids and their derivatives or unsaturated fatty acids such as palmitoleic, oleic, erucic, nervonic, linoleic, α -linolenic, γ -linolenic, di-homo- γ -linolenic, arachidonic, eicosapentaenoic, docosahexanoic acids and their derivatives.

Preferably, the fatty acids are produced by the hydrolysis of triglycerides present in vegetable and animal oils such as copra, palm, olive, peanut, rapeseed, sunflower, soy, cotton, linseed, beef tallow. Natural oils may have been genetically modified in order to enrich the content thereof of certain fatty acids, e.g. rapeseed oil or oleic sunflower oil.

In general, the fatty amine of formula (I) according to the invention is preferably obtained from natural plant or animal resources. The processing allows producing fatty amines from natural oils that can lead to a mixture of primary, secondary and tertiary polyamines.

According to a particular embodiment of the invention, when a plurality of fatty amines of formula (I) are used to prevent and/or reduce metallic losses of parts of an engine, said fatty amines form a mixture of fatty amines comprising in variable proportions, all or a portion of compounds meeting the following formulae (Ia), (Ib) and (Ic):



wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently a hydrogen atom, or a saturated or unsaturated, linear or branched alkyl moiety, comprising between 1 and 22 carbon atoms; or a $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,
- R_3 represents a saturated or unsaturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms,

The preferences and advantages for the definitions of the moieties R_1 , R_2 , R_3 , R_4 , R_5 and R_6 of fatty amines of formulae (Ia), (Ib) and (Ic) are such as defined hereinabove for the fatty amine of formula (I) according to the invention.

According to another particular embodiment, the mixture of fatty amines of formula (I) is in a purified form, i.e. mainly comprising a single type of amine of formula (Ia), (Ib) or (Ic), e.g. mainly diamines of formula (Ia), triamines of formula (Ib) or further mainly tetramines of formula (Ic). In particular, the mixture of fatty amines comprises mainly tetramines of formula (Ic).

According to a particular embodiment of the invention, the mixture of fatty amines of formula (I) comprises mainly:

- diamines of formula (Ia), or
- triamines of formula (Ib), or
- tetramines of formula (Ic),

wherein the moieties R_1 , R_2 , R_3 , R_4 , R_5 and R_6 are such as defined hereinabove.

In another embodiment of the invention, the mixture of fatty amines of formula (I) mainly comprises diamines of formula (Ia) wherein:

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 20 carbon atoms, preferably from 14 to 20 carbon atoms, more preferably from 16 to 20 carbon atoms,
- R_2 represents a saturated, linear or branched alkyl moiety comprising from 1 to 5 carbon atoms, preferably from 1 to 3 carbon atoms, more preferably a methyl moiety,
- R_3 represents an ethyl or propyl moiety,
- R_4 and R_5 represent independently a saturated, linear or branched moiety comprising from 1 to 5 carbon atoms, preferably from 1 to 3 carbon atoms, more preferably a methyl moiety.

Preferably, the mixture of fatty amines of formula (I) mainly comprises diamines of formula $R_1-[(NR_2)-R_3]-NH_2$ (IIa), triamines of formula $R_1-[(NR_2)-R_3]_2-NH_2$ (IIb), or tetramines of formula $R_1-[(NR_2)-R_3]_3-NH_2$ (IIc), wherein:

- R_1 or R_2 represent at least a saturated or unsaturated alkyl moiety, obtained from a fatty acid coming from tallow or soy oil, or coconut oil or (oleic) sunflower oil, and
- R_3 represents a saturated or unsaturated, linear or branched alkyl moiety comprising at least 2 carbon atoms.

As defined by the invention, when R_1 or R_2 represent a saturated alkyl moiety, said saturated alkyl moiety is obtained from a saturated fatty acid or from an unsaturated fatty acid having undergone hydrogenation, in particular of all of the double bonds thereof.

Advantageously, the mixture of fatty amines of formula (I) mainly comprising tetramines of formula $R_1-[(NR_2)-R_3]_3-NH_2$ (IIc) are in the form:

- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 16 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms
- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising at least 18 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms, and
- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising at least 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms.

In particular, the mixture of fatty amines of formula (I) mainly comprising tetramines of formula $R_1-[(NR_2)-R_3]_3-NH_2$ (IIc) are in the form:

- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising from 14 to 16 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms,
- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising at least 18 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms, and
- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated or unsaturated, linear or branched alkyl moiety comprising at least 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms,

the sum of the concentration by weight of said fatty amines of formula (IIc) being greater than 90% with respect to the weight of said mixture of fatty amines.

Advantageously, the mixture of fatty amines of formula (I) mainly comprising tetramines of formula $R_1-[(NR_2)-R_3]_3-NH_2$ (IIc) are also in the form:

- of at least one fatty amine of formula (IIc) wherein R_1 represents an unsaturated, linear or branched alkyl moiety comprising from 16 to 20 carbon atoms, preferably

from 18 to 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms and

- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated, linear or branched alkyl moiety comprising from 16 to 20 carbon atoms, preferably from 18 to 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms.

In particular, the mixture of fatty amines of formula (I) mainly comprising tetramines of formula $R_1-[(NR_2)-R_3]_3-NH_2$ (IIc) are in the form:

- of at least one fatty amine of formula (IIc) wherein R_1 represents an unsaturated, linear or branched alkyl moiety comprising from 16 to 20 carbon atoms, preferably from 18 to 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms,
- of at least one fatty amine of formula (IIc) wherein R_1 represents a saturated, linear or branched alkyl moiety comprising from 16 to 20 carbon atoms, preferably from 18 to 20 carbon atoms; R_2 represents a hydrogen atom; and R_3 represents a saturated, linear alkyl moiety, comprising from 2 to 6 carbon atoms,

the sum of the concentration by weight of said fatty amines of formula (IIc) being greater than 90% with respect to the weight of said mixture of fatty amines.

Preferably, the mixture of fatty amines of formula (I) does not comprise fatty amines other than fatty acids meeting formula (IIc).

According to a particular embodiment of the invention, when a single fatty amine of formula (I) is used to prevent and/or reduce metallic losses of parts of an engine, said fatty amine meets one of the following formulae:

- a diamine of formula (IIa), or
- a triamine of formula (IIb), or
- a tetramine of formula (IIc),

wherein,

- R_1 represents a saturated, linear or branched hydrocarbon moiety comprising from 14 to 22 carbon atoms,
- R_2 represents independently a hydrogen atom or a saturated, linear or branched hydrocarbon moiety comprising between 14 and 22 carbon atoms,
- R_3 represents a saturated, linear hydrocarbon moiety comprising between 2 and 6 carbon atoms.

In this embodiment, the fatty amine of formula (I) is preferably a tetramine of formula (IIc) wherein,

- R₁ represents a saturated, linear or branched alkyl moiety comprising between 14 and 18 carbon atoms,
- R₂ represents independently a hydrogen atom or a saturated, linear or branched hydrocarbon moiety comprising between 14 and 18 carbon atoms,
- R₃ represents a saturated, linear hydrocarbon moiety comprising between 2 and 6 carbon atoms.

In this embodiment, the fatty amine of formula (I) is advantageously a tetramine of formula (IIc) wherein,

- R₁ represents a saturated, linear or branched alkyl moiety comprising between 16 and 18 carbon atoms,
- R₂ represents independently a hydrogen atom or a saturated, linear or branched hydrocarbon moiety comprising between 16 and 18 carbon atoms,
- R₃ represents an ethyl or propyl moiety.

Preferably, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the mixtures of polyalkylamines comprise at least 5% by weight of compounds having a pure linear structure, given that said compounds are revealed to have an acceptable viscosity profile.

According to one embodiment, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the mixtures of polyalkylamines comprise at least 4% by weight (%w/w), preferably at least 5%w/w, preferably at least 6%w/w, preferably more than 7%w/w, preferably more than 7.5%w/w, preferably more than 10%w/w, preferably more than 20%w/w of branched compounds of which at least one of n or z is greater than or equal to 1.

For products with formulae (III), it means that for branched products, n must be greater than or equal to 1.

Preferably, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), when n, o, p or z is equal to 0, the hydrogen atom present at the end of the chain is covalently bound to the corresponding secondary nitrogen atom.

Preferably, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the mixture comprises compounds of formulae (III) and/or (IV), wherein n, o,

p and z are different from 0, are equal to 1 or 2, preferably when n, o, p and z are different from 0, they are equal to 1.

According to a preferred embodiment, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the mixture essentially comprises compounds of formulae (III) and/or (IV), for which n, o, p and z independently represent 0, 1 or 2, preferably n, o, p and z independently represent 0 or 1.

According to a preferred embodiment, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the mixture comprises essentially compounds of formulae (III) and/or (IV), and their derivatives for which n, o, p and z independently represent 0, 1 or 2, preferably n, o, p and z independently represent 0 or 1.

The derivatives of compounds of formulae (III) and/or (IV) are described below.

According to a preferred embodiment, each group R is, independently of each other, a linear or branched alkyl group comprising from 14 to 22 carbon atoms, preferably 14 to 20 carbon atoms, preferably from 16 to 20 carbon atoms.

In general, fatty amines of formula (III) and (IV) according to the invention are mainly obtained from carboxylic acids. These acids are dehydrated in the presence of ammonia for yielding nitriles and then undergo a catalytic hydrogenation in particular for leading to fatty amines.

Fatty amines of formula (III) and (IV) used according to the invention are obtained from at least one carboxylic acid, preferably from at least a fatty acid.

The alkyl group of the fatty alkyl amines of formula (III) and (IV) used according to the invention have a number of carbon atoms corresponding to the number of carbon atoms of the carbon chain of the carboxylic acid, preferably corresponding to the number of carbon atoms of the carbon chain of the fatty acid.

As defined by the invention, a same fatty amine of formula (I) can be substituted by a plurality of alkyl moieties obtained from a plurality of carboxylic acids, identical or different, preferably obtained from a plurality of fatty acids, identical or different.

As defined by the invention, a same fatty amine of formula (I) can be substituted by a plurality of alkyl moieties obtained from a plurality of carboxylic acids, identical or different, preferably obtained from a plurality of fatty acids, identical or different.

While the two R groups could be different, said groups are, according to a preferred embodiment, identical, such compounds being produced more economically. Independently of the fact of whether or not said groups are identical, one or both R groups independently, come from chemical or natural raw material such as oils or natural

greases. In particular, if a natural raw material is used, this means that each R group could have a particular distribution over the length of the carbon chain. Appropriately, R is derived from oil or grease of animal or plant origin such as tallow, coconut oil and palm oil. Since the preparation of the fatty di-alkyl polyalkylamines according to the invention comprises a hydrogenation stage, it may be advantageous, during the method of preparation of the products of the invention, to use hydrogenated R groups. Advantageously, the R group is a hydrogenated tallow group. Preferably, the R group of the raw material is unsaturated and is (partially) hydrogenated during the method of preparation of the fatty polyalkylamine.

According to a particular embodiment of the invention, when the fatty amine is a mixture of polyalkylamines of formulae (III) and/or (IV), the alkyl group R is obtained from the fatty acid chosen amongst the caprylic, pelargonic, capric, undecylenic, lauric, tridecylenic, myristic, pentadecylic, palmitic, margaric, stearic, nonadecylic, arachic, heneicosanoic, behenic, tricosanoic, lignoceric, pentacosanoic, cerotic, heptacosanoic, montanic, nonacosanoic, melissic, hentriacontanoic, laceroic acids and their derivatives or unsaturated fatty acids such as palmitoleic, oleic, erucic, nervonic, linoleic, α -linolenic, c-linolenic, di-homo-c-linolenic, arachidonic, eicosapentaenoic, docosahexanoic acids and their derivatives.

Preferably, the fatty acids are produced by the hydrolysis of triglycerides present in vegetable and animal oils such as copra, palm, olive, peanut, rapeseed, sunflower, soy, cotton, linseed, beef tallow. Natural oils may have been genetically modified in order to enrich the content thereof of certain fatty acids, e.g. rapeseed oil or oleic sunflower oil.

The compositions of fatty di-alkyl polyalkylamine derivatives of formulae (III) and/or (IV) according to the invention comprise compounds for which one or a plurality of NH fragments of the fatty polyalkylamine of the invention are methyl, alkoxylys or both. It has been discovered that such compounds have an advantageous solubility, in particular in lubricant oils. Advantageously, the alkoxyyl derivatives are butoxylys, propoxylys and/or ethoxylys. If two or more alkoxylyng agents are used, the same can be used in any order, e.g. EO-PO-EO and the different alkoxy units can be of the polyedric type and/or present randomly. Advantageously, an $-NH_2$ group is alkoxylyated classically with one or a plurality of alkylene oxides to form the group $-NH-AO-H$, where AO represents one or a plurality of alkylene-oxy moieties. Moreover, the $-NH-AO-H$ group can be obtained alkoxylyated to form $-N(AO-H)_2$ moieties. In particular, when large quantities of alkylene oxide (i.e. more than 8 moles of alkylene oxides per mole of polyalkylamine) are used, in general one or a plurality of secondary amines, if present, are alkoxylyated.

According to one embodiment, the primary and secondary amine functional groups of the di-alkyl polyamine of formulae (III) and/or (IV) are alkoxyated. According to another embodiment, the fatty di-alkyl polyalkylamines are derivatized by methylation of one or a plurality of NH functional groups in a manner known to a person skilled in the art, e.g. by reaction with formic acid or formaldehyde. According to one embodiment, one or a plurality of OH functional groups of the alkoxyated fatty di-alkyl polyalkylamines are methylated conventionally.

However, since it may be more economical to prepare the mixture of polyalkylamines of formula (IV), mixtures of polyalkylamine of formula (IV) are preferred. If appropriate, mixtures of polyalkylamine of formulae (III) and/or (IV) are used.

The branched polyalkylamines of the invention can be produced by any synthesis path known to a person skilled in the art. A conventional method of production could be from a diamine and involve two or a plurality of cycles, preferably two for economic reasons, each cycle comprising a cyanoethylation stage and a hydrogenation stage. In the following, this method is called a two-stage method. In an alternative method, an equivalent of di-alkyl diamine can react in a single stage with two or more equivalents of acrylonitrile followed by hydrogenation. In this case, optional additional cycles involving cyanoethylation and hydrogenation stages can be envisaged. Such a single-stage method can be advantageous since the same require fewer intermediate stages. In order to increase the branching in the two-stage method, an acid catalyst is used such as HCl or acetic acid. Moreover, the increase in temperature of the reaction during the cyanoethylation also allows the branching in this method to be increased. During the execution of a multi-cycle method, the temperature of a later stage of cyanoethylation is higher than the temperature of a preceding cyanoethylation stage, allowing a compound with the desired branching to be obtained. According to one embodiment, more than one mole of acrylonitrile per mole of initial polyamine is used also allowing the branching of the expected product to be increased to the desired level. A solvent is used appropriately and in order to maintain a homogeneous reaction mixture. Preferred solvents comprise C₁₋₄ alcohols and C₂₋₄ diols. Preferably, ethanol is used as it allows for particular ease of use. Surprisingly, it has been shown that C₁₋₄ alcohols and C₂₋₄ diols are not simple solvents but also possess co-catalytic activity during the cyanoethylation stage. The quantity of solvent used can vary over a large range. For economic reasons, preferably a minimum quantity is used. The quantity of solvent, in particular during the cyanoethylation stage, is preferably less than 50, 40, 30 or 25% by weight with respect to the liquid reaction mixture. The quantity of solvent, in particular during the cyanoethylation stage,

is preferably greater than 0.1, 0.5, 1, 5, or 10% by weight with respect to the liquid reaction mixture.

According to one embodiment, the mixture of fatty di-alkyl polyalkylamines of formulae (III) and/or (IV) according to the invention is characterized by a BN measured according to the standard ASTM D-2896 comprised between 150 and 350 mg KOH/g of amine, preferably between 170 and 340 and even more preferably between 180 and 320.

According to one embodiment, in cylinder lubricants according to the invention, the percentage by weight of the mixture of di-alkyl polyalkylamine with respect to the total weight of the lubricant composition is selected so that the BN furnished by said compounds represents a contribution comprising between 5 and 60 mg KOH/g of lubricant, preferably between 10 and 30 mg KOH/g of lubricant, said cylinder lubricant BN measured according to the standard ASTM D-2896.

Lubricant composition

The fatty amine of formula (I) or the mixture of fatty amines of formulae (III) and/or (IV) or the mixture of fatty amines of formulae (I), (III) and/or (IV) according to the invention allows metallic losses of parts of an engine, preferably a marine engine to be prevented and/or reduced, is present in a lubricant composition. Said lubricant composition comprises:

- at least one base oil, preferably a lubricating base oil for a marine engine,
- at least one detergent containing alkali metals or alkaline-earth metals overbased by metal carbonate salts.

Preferably, the lubricant composition has a BN determined as per the standard ASTM D-2896, of less than or equal to 15 milligrams of potash per gram of lubricant, more preferably less than or equal to 40 milligrams.

Advantageously, the lubricant composition has a BN determined according to the standard ASTM D-2896, comprised between 40 and 120 milligrams of potash per gram of lubricant, preferably between 50 and 100 milligrams of potash per gram of lubricant.

Advantageously, the lubricating composition also has a BN determined as per the standard ASTM D-2896, comprised between 15 and 40 milligrams of potash per gram of lubricant, preferably between 20 and 40 milligrams of potash per gram of lubricant.

According to one embodiment of the invention, the weight percentage of fatty amine with respect to the total weight of cylinder lubricant is chosen so that the BN furnished by the fatty amine represents a contribution of at least 2 milligrams of potash

per gram of lubricant with respect to the total BN of said cylinder lubricant, preferably at least 5 milligrams of potash per gram of lubricant to the total BN of said cylinder lubricant.

The part of the BN brought by a fatty amine in the cylinder lubricant according to the invention (in milligrams of potash per gram of final lubricant or again BN "points") is calculated from its intrinsic BN measured according to the standard ASTM D-2896 and its weight percentage in the final lubricant:

$$\text{BN amine lub} = x \cdot \text{BN amine} / 100$$

$$\text{BN amine lub} = \text{contribution of the amine to the BN of the final lubricant}$$

$$x = \% \text{ by weight of amine in the final lubricant}$$

$$\text{BN amine} = \text{intrinsic BN of the amine alone (ASTM D-2896)}.$$

According to one embodiment of the invention, the weight percentage of fatty amine of formulae (I), (III) and/or (IV) with respect to the total weight of cylinder lubricant is chosen so that the BN furnished by the fatty amine represents a contribution from 2 to 30 milligrams of potash per gram of lubricant, more preferably from 5 to 25 milligrams of potash per gram of lubricant to the total BN of said cylinder lubricant.

According to a preferred embodiment of the invention, the fatty amine of formula (I) or the mixture of fatty amines of formulae (III) and/or (IV) or the mixture of fatty amines of formulae (I), (III) and/or (IV) is added in an amount of from 0.1 to 15%, preferably from 0.5 to 10%, preferably from 0.5 to 8% or from 3 to 10% by weight with respect to the total weight of the lubricant composition.

In another embodiment of the invention, the fatty amine of formula (I) represents 0.5 to 10%, preferably 0.5 to 8% by weight with respect to the total weight of the lubricant composition.

In another embodiment of the invention, the percentage by weight of the mixture of di-alkyl polyalkylamine of formulae (III) and/or (IV) with respect to the total weight of lubricant is comprised between 0.1 and 15%, preferably between 0.5 and 10%, advantageously between 3 and 10%.

Preferably, the lubricant composition further comprises at least one neutral detergent.

With regard to detergents used in the lubricant compositions according to the present invention, the same are well known to a person skilled in the art.

According to a particular embodiment of the invention, the detergents commonly used in the formulation of lubricant compositions are typically anionic compounds comprising a long lipophilic hydrocarbon chain and a hydrophilic head. The associated cation is typically a metal cation of an alkali or alkaline earth metal.

The detergents are preferentially chosen from alkali metal or alkaline earth metal salts of carboxylic acids, sulfonates, salicylates and naphthenates, as well as the phenate salts.

The alkali metals and alkaline earth metals are preferentially calcium, magnesium, sodium or barium.

Said metal salts can contain the metal in an approximately stoichiometric amount. In such case, one refers to non-overbased or "neutral" detergents, although the same also furnish a certain basicity. Such "neutral" detergents typically have a BN, measured according to ASTM D2896, of less than 150 mg KOH/g, or less than 100, or else less than 80 mg KOH/g.

Such so-called neutral detergents could contribute in part to the BN of the lubricants according to the present invention. Neutral detergents such as carboxylates, sulfonates, salicylates, phenates, naphthenates of alkali metals and alkaline earth metals, e.g. calcium, sodium, magnesium or barium type, will be used.

When the metal is in excess (in a quantity greater than the stoichiometric quantity), one is dealing with so-called overbased detergents. The BN thereof is high, greater than 150 mg KOH/g, typically comprising between 200 and 700 mg KOH/g, generally comprised between 250 and 450 mg KOH/g.

The excess metal that furnishes the overbased character to the detergent is in the form of metal salts insoluble in oil, e.g. carbonate, hydroxide, oxalate, acetate, glutamate, preferentially carbonate.

In the same overbased detergent, the metals of said insoluble salts can be either the same as the metals of oil-soluble detergents or different. The metals are preferentially selected from calcium, magnesium, sodium or barium.

Thereby, the overbased detergents are in the form of micelles composed of insoluble metal salts held in suspension in the lubricant composition by the detergents in the form of oil-soluble metal salts.

Said micelles can contain one or a plurality of types of insoluble metal salts, stabilized by one or a plurality of types of detergent.

Overbased detergents comprising a single type of soluble metal salt detergent will generally be named after the nature of the hydrophobic chain of said latter detergent.

Therefore, the detergents will be referred to as a carboxylate, phenate, salicylate, or naphthenate type depending on whether the detergent is respectively a carboxylate, a phenate, a salicylate, a sulfonate or a naphthenate.

Overbased detergents will be referred to as mixed type if the micelles comprise a plurality of types of detergents, different from each other by the nature of the hydrophobic chain thereof.

For use in the lubricant compositions according to the present invention, the oil-soluble metal salts will preferentially be carboxylates, phenates, sulfonates, salicylates, and mixed phenate-sulfonate and/or salicylates detergents of calcium, magnesium, sodium or barium.

The insoluble metal salts furnishing the overbased character are alkali metal and alkaline earth metal carbonates, preferentially calcium carbonate.

The overbased detergents used in the lubricant compositions according to the present invention will preferentially be carboxylates, phenates, sulfonates, salicylates and mixed phenate-sulfonate-salicylate detergents overbased with calcium carbonate.

According to a particular embodiment of the invention, the base oil comprised in the lubricant composition is chosen from mineral, synthetic or vegetable oils, as well as mixtures thereof.

The mineral or synthetic oils generally used in the application belong to one of the classes defined in the API classification as summarized in the table below.

	Concentration of saturates	Concentration of sulfur	Viscosity index
Group 1 Mineral oils	< 90%	> 0.03%	$80 \leq VI < 120$
Group 2 Hydrocracked oils	$\geq 90\%$	$\leq 0.03\%$	$80 \leq VI < 120$
Group 3 Hydro-isomerized oils	$\geq 90\%$	$\leq 0.03\%$	≥ 120
Group 4	PAO		
Group 5	Other bases not included in bases of groups 1 to 4		

Group 1 mineral oils can be obtained by distillation of chosen naphthenic or paraffinic crude oils followed by purification of said distillates by methods such as solvent extraction, solvent or catalytic dewaxing, hydrotreatment or hydrogenation.

Group 2 and 3 oils are obtained by more demanding purification methods, e.g. a combination of hydrotreatment, hydrocracking, hydrogenation and catalytic dewaxing.

Examples of Group 4 and 5 synthetic base oils include poly-alpha olefins such as polybutenes, polyisobutenes, alkylbenzenes.

Said base oils can be used alone or as a mixture. A mineral oil can be combined with a synthetic oil.

Cylinder oils for marine diesel 2-stroke engines typically have a viscosity grade of SAE-40 to SAE-60, generally SAE-50 is equivalent to a kinematic viscosity at 100°C comprised between 16.3 and 21.9 mm²/s.

Grade 40 oils have a kinematic viscosity at 100°C comprised between 12.5 and 16.3 mm²/s.

Grade 50 oils have a kinematic viscosity at 100°C comprised between 16.3 and 21.9 mm²/s.

Grade 60 oils have a kinematic viscosity at 100°C comprised between 21.9 and 26.1 V.

According to the uses of the profession, it is preferred to formulate cylinder oils for marine diesel two-stroke engines having a kinematic viscosity at 100°C comprised between 18 and 21.5, preferably between 19 and 21.5 mm²/s.

Said viscosity can be obtained by mixing additives and base oils e.g. containing Group 1 mineral bases such as Neutral Solvent bases (e.g. 500NS or 600 NS) and Brightstock. Any other combination of mineral, synthetic or vegetable bases having, in a mixture with the additives, a viscosity compatible with grade SAE-50 can be used.

Typically, a conventional formulation of cylinder lubricant for slow 2-stroke marine diesel engines is of grade SAE-40 to SAE-60, preferentially SAE-50 (according to the SAE J300 classification) and comprises at least 50% by weight of a lubricant base oil of mineral and/or synthetic origin, suitable for use in marine engines, e.g. API Group 1 i.e. obtained by distillation of chosen crude oils followed by purification of said distillates by methods such as solvent extraction, solvent or catalytic dewaxing, hydrotreatment or hydrogenation. The Viscosity Index (VI) thereof is comprised between 80 and 120; the concentration of sulfur thereof is greater than 0.03 % and the concentration of saturates thereof is less than 90 %.

According to a particular embodiment of the invention, the lubricant composition may further comprise one or a plurality of thickening additives the role of which is to increase the viscosity of the composition, both hot and cold, or of additives improving the Viscosity Index (VI).

Preferably, said additives are most often polymers with low molecular weight, on the order of 2,000 to 50,000 dalton (Mn).

The additives can be chosen from PIBs (on the order of 2,000 dalton), Polyacrylate or Polymethacrylates (on the order of 30,000 dalton), Olefin copolymers, Copolymers of olefin and Alpha Olefins, EPDM, Polybutenes, Poly-alphaolefins with high molecular weight (viscosity 100°C > 150), Styrene-Olefin copolymers, whether hydrogenated or not.

According to a particular embodiment of the invention, the base oil(s) comprised in the lubricant composition according to the invention can be partially or totally substituted by said additives.

Henceforth, the polymers used to partially or totally substitute one or a plurality of the base oils are preferentially the abovementioned thickeners such as PIB type (e.g. marketed under the name of Indopol H2100).

According to a particular embodiment of the invention, the lubricant composition can further comprise at least one anti-wear additive.

Preferably, the anti-wear additive is Zinc di-thiophosphate or DTPZn. Various phosphorus, sulfur, nitrogen, chlorine and boron compounds are also found in said category.

There is a wide variety of anti-wear additives, but the most widely used category is that of phosphor-sulfur additives such as metal alkylthiophosphates, in particular Zinc alkylthiophosphates, and more specifically Zinc dialkyldithiophosphates or DTPZn.

Amine phosphates, polysulfides, in particular sulfur olefins, are also commonly used anti-wear additives.

Anti-wear and extreme pressure additives such as nitrogen and sulfur additives are also usually found in lubricant compositions, such as e.g. metal dithiocarbamates, in particular molybdenum dithiocarbamate. Glycerol esters are also anti-wear additives. Examples include e.g. of mono-, di- and trioleates, monopalmitates and monomyristates.

According to a particular embodiment of the invention, the lubricant composition can further comprise at least one dispersant.

Dispersants are well known additives used in the formulation of lubricant compositions, in particular for application in the marine field. The primary role thereof is to maintain in suspension the particles initially present or appearing in the lubricant composition during the use thereof in the engine. Dispersants prevent the agglomeration of particles by playing on steric congestion. Dispersants can also have a synergistic effect on neutralization.

The dispersants used as lubricant additives typically contain a polar group, associated with a relatively long hydrocarbon chain, generally containing from 50 to 400 carbon atoms. The polar group typically contains at least one nitrogen, oxygen or phosphorus element.

Derivative compounds of succinic acid are dispersants particularly used as lubricant additives. Succinimides, obtained by condensation of succinic anhydrides and

amines, succinic esters obtained by condensation of succinic anhydrides and alcohols or polyols, are used in particular.

Said compounds can then be treated with various compounds, in particular sulfur, oxygen, formaldehyde, carboxylic acids and compounds containing boron or zinc for producing e.g. borated succinimides or zinc-blocked succinimides.

Mannich bases, obtained by polycondensation of alkyl-substituted phenols, formaldehyde and primary or secondary amines, are also compounds used as dispersants in lubricants.

It is possible to use a dispersant of the family of PIB succinimides e.g. borated or blocked with zinc.

According to a particular embodiment of the invention, the lubricant composition can further comprise any type of functional additives suitable for the use thereof, e.g. anti-foam additives, to counter the effect of detergents, which can be e.g. polar polymers such as polymethylsiloxanes, polyacrylates, antioxidant and/or rust-inhibiting additives, e.g. organo-metallic detergents or thiadiazoles. The same are known to a person skilled in the art.

According to the present invention, the compositions of the lubricants described refer to the compounds taken separately before mixing, it being understood that said compounds may or may not retain the same chemical form before and after mixing. Preferably, the lubricants according to the present invention obtained by mixing the compounds taken separately are not in emulsion nor in microemulsion form.

Engine

The use of one or a plurality of fatty amines of formulae (I), (III) and/or (IV) according to the invention soluble in a lubricant composition allows metallic losses of parts of an engine to be prevented and/or reduced.

According to a particular embodiment of the invention, the use of one or a plurality of fatty amines of formulae (I), (III) and/or (IV) allows metallic losses of parts of a two-stroke, marine engine to be prevented and/or reduced during the combustion of fuel oil.

According to a particular embodiment of the invention, the use of one or a plurality of fatty amines of formulae (I), (III) and/or (IV) according to the invention allows metallic losses of parts in the hot zones, in particular the piston-ring jacket zone, of a two-stroke or four-stroke, marine engine to be prevented and/or reduced during the combustion of fuel oil.

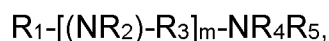
According to a particular embodiment of the invention, the fuel oil has a sulfur concentration less than 3.5% by weight with respect to the total weight of the fuel oil.

The different embodiments, the variants, the preferences and the advantages described hereinabove can be taken separately or in combination for the implementation of the first subject matter of the invention.

Process

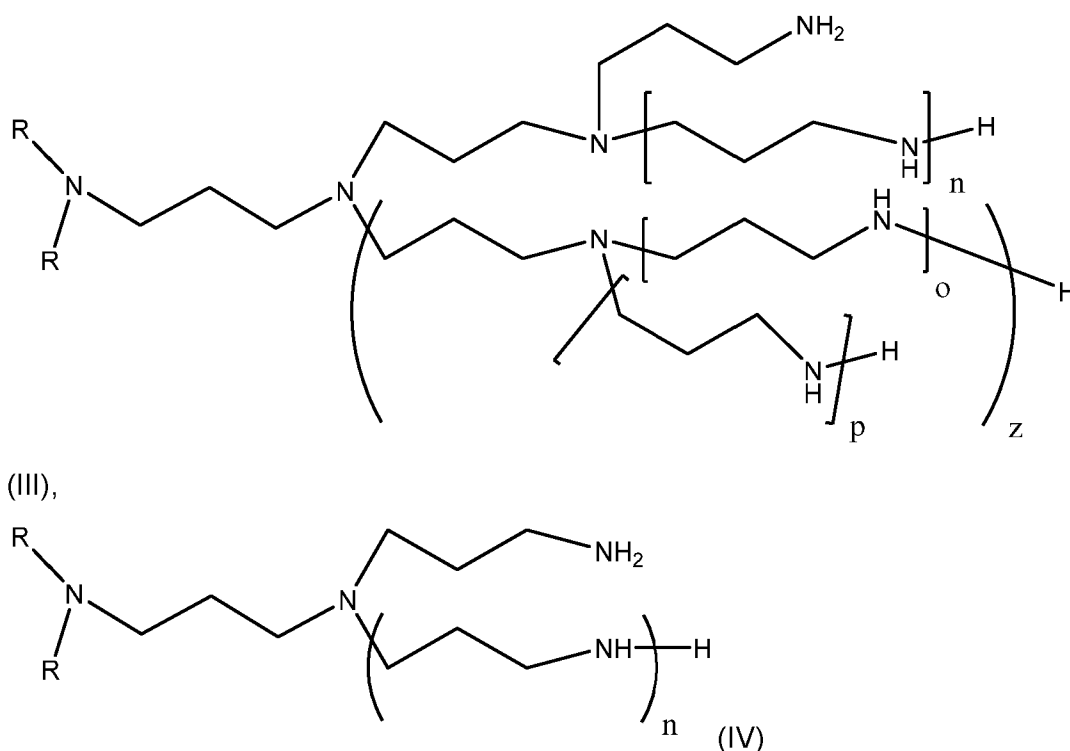
A further subject matter of the invention is a method for preventing and/or reducing metallic losses of parts of an engine, in particular a marine engine wherein said parts are put in contact with one or a plurality of soluble fatty amines in a lubricant composition, the fatty amine being chosen from:

- compounds with the formula (I):



wherein,

- R_1 represents a saturated or unsaturated, linear or branched hydrocarbon moiety comprising from 12 to 22 carbon atoms,
- R_2 , R_4 or R_5 represent independently a hydrogen atom, or a saturated or unsaturated, linear or branched alkyl moiety, comprising between 1 and 22 carbon atoms; or an $(R_6-O)_q-H$ moiety wherein R_6 is a saturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms, and q represents a whole number greater than or equal to 1,
- R_3 represents a saturated or unsaturated, linear or branched alkyl moiety comprising between 2 and 6 carbon atoms,
- m is a whole number comprised between 1 and 10, more preferably between 1 and 6, again more preferably is chosen amongst 1, 2 or 3,
- a mixture of fatty polyalkylamines comprising one or a plurality of polyalkylamines with the formulae (III) and/or (IV):



wherein

- R, identical or different represents a linear or branched alkyl group comprising from 8 to 22 carbon atoms,
- n and z, independently of each other, represent 0, 1, 2 or 3, and
- when z is greater than 0, o and p represent independently of each other, 0, 1, 2 or 3,

said mixture comprising at least 3% by weight of branched compounds such that at least one of n or z is greater than or equal to 1, or of their derivatives, and

- a mixture of fatty amines of formulae (I), (III) and/or (IV).

The different embodiments, preferences, advantages, variants described hereinabove covering the use of one or a plurality of soluble, fatty amines in a lubricant composition for preventing and/or reducing metallic losses of parts of an engine, preferably a marine engine, apply separately or in combination to the second subject matter of the invention covering the method described hereinabove.

The invention is illustrated by the following examples, but is not limited to them.

Experimental protocol for measurements of metallic losses from a metallic part

200 g of the lubricant composition comprising one or a plurality of fatty amines according to the invention is introduced into a 700 ml test vessel and said test vessel is heated to 60°C while being stirred strongly for a period of 30 minutes.

A cast iron plate, sanded, cleaned and weighed beforehand, is immersed in the test vessel and thus heated and stirred, and then an amount of sulfuric acid, diluted to 50% so as to neutralize all or part of the total BN of said lubricant composition, was added progressively over 1 hour and 30 minutes. The amount of sulfuric acid diluted to 50% and added into the lubricant composition is calculated according to the maximum BN number needed to neutralize. The addition flow-rate of sulfuric acid diluted to 50% is calculated according to the total amount of sulfuric acid to add over a period of 1h30.

The lubricant composition thus acidified and comprising the plate was then stirred for an additional 30 minutes in order to ensure that the BN neutralization reaction was terminated.

The plate immersed in the acidified lubricant composition is then withdrawn from the test vessel, then weighed in order to determine the metallic losses from said cast iron plate due to sulfuric acid attack.

EXAMPLE

Evaluation of metallic losses from a metallic part put in contact with fatty amines according to the invention and with sulfuric acid

It concerns assessing metallic losses from a metallic part put directly in contact with sulfuric acid and with fatty amines according to the invention contained in a lubricant composition.

For this, different lubricant compositions were prepared from the following compounds:

- a base lubricating oil comprising a mixture of mineral oils from group I and/or II, in particular Brightstock type oils,
- a detergent package,
- a mixture of fatty amines 1 comprising mainly polyalkylamines of formulae (III) and/or (IV),
- a fatty amine mixture 2 comprising mainly tetramines of formula (I),
- a fatty amine mixture 3 comprising mainly diamines of formula (I),
- a fatty amine mixture 4 comprising mainly triamines of formula (I),
- a fatty amine mixture 5 comprising mainly tetramines of formula (I).

Compositions L₁ to L₇ according to the invention are described in table I as well as a lubricant composition L₈ only comprising a base oil and a detergent package; the percentages indicated correspond to percentages by weight.

Table I

Compositions	L ₁	L ₂	L ₃	L ₄	L ₅	L ₆	L ₇	L ₈ (standard)	L ₉	L ₁₀	L ₁₁
Base oil	74.5	76	67.7	63.9	68.7	60.1	60.3	63	68	69	69.5
Detergent package	22.1	20.6	26.8	28.6	26.7	35	34.8	37	26.8	26.8	26.8
Fatty amine 1	-	-	5.5	7.5	4.6	4.9	4.9	-	-	-	-
Fatty amine 2	3.4	3.4	-	-	-	-	-	-	-	-	-
Fatty amine 3	-	-	-	-	-	-	-	-	5.2	-	-
Fatty amine 4	-	-	-	-	-	-	-	-	-	4.2	-
Fatty amine 5	-	-	-	-	-	-	-	-	-	-	3.7

The results obtained relating to metallic losses of parts put in contact with sulfuric acid and with the lubricant compositions L₁ to L₈ respectively are described in table II.

Table II

Compositions	L ₁	L ₂	L ₃	L ₄	L ₅	L ₆	L ₇	L ₈ (standard)	L ₉	L ₁₀	L ₁₁
Metallic losses	1.8	1.4	1.55	0.15	0	0	0.55	89	1	2.8	3.3

It is observed that the presence of fatty amines according to the invention contained in the compositions L₁ to L₇ and L₉ to L₁₁ allow the metallic losses from a part put directly in contact with sulfuric acid to be significantly decreased or even avoided and this independently of the concentration of fatty acids in the lubricant composition, unlike the standard composition which did not comprise fatty amines according to the invention. In particular, the metallic losses of parts did not exceed 4 mg when said parts were immersed in an acid medium and in the presence of different types of fatty amines respectively according to the invention contained in lubricant compositions unlike the metallic losses of parts put directly in contact with sulfuric acid and a lubricant composition not comprising fatty amines according to the invention that exceeded 80 mg.

It also relates to assessing metallic losses from a metallic part put directly in contact with fatty amines according to the invention contained in a lubricant composition and with a large excess of sulfuric acid in order to demonstrate that the reduction of metallic losses under such conditions was not due to the neutralization of the sulfuric acid by said fatty amines but indeed came from the passivation of all or part of the surface of the metallic parts by said fatty amines.

To do this, two metallic parts were immersed respectively in lubricant compositions L₄ and L₈ in the presence of a large excess of sulfuric acid. The results obtained relating to metallic losses of parts put in contact with a large excess of sulfuric acid and with the lubricant compositions L₄ and L₈ respectively are described in table III.

The test allowing assessments to be made of metallic losses from a metallic part put directly in contact with fatty amines according to the invention contained in a lubricant composition and with a large excess of sulfuric acid was performed according to the experimental protocol for measurements of metallic losses from a metallic part described hereinabove. In this test, the amount of sulfuric acid diluted to 50% introduced into the lubricant composition according to the invention was calculated so as to neutralize 150% of the total BN of the lubricant composition

Table III

Compositions	L ₄	L ₈ (standard)
Metallic losses (mg)	7	117.7

It was observed that even in the presence of a large excess of sulfuric acid, the metallic part lost little material; i.e. said part lost 7 mg when in contact with the fatty amines according to the invention contained in a lubricant composition, unlike a metallic part put in contact with a lubricant composition not comprising fatty amines according to the invention for which the metallic losses went beyond 115 mg.

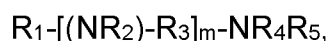
From now on, it is clearly demonstrated that the use of at least one fatty amine and/or fatty amine derivative contained in a lubricant composition allows metallic losses from a part of an engine, in particular a marine engine, i.e. in contact with an acid medium, to be significantly reduced or even avoided.

ANVENDELSE AF EN FEDTAMIN TIL FOREBYGGELSE OG/ELLER REDUKTION AF METALTAB PÅ DELE I EN MOTOR

PATENTKRAV

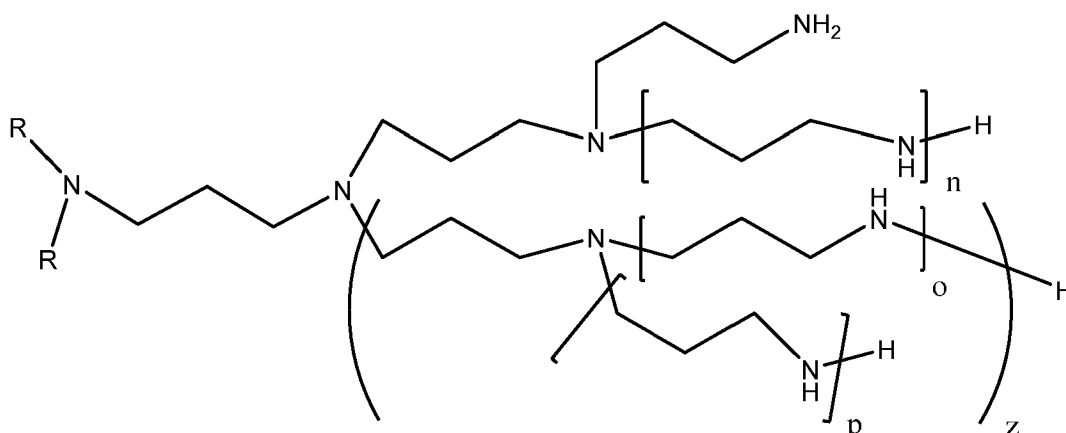
1. Anvendelse af en eller flere fedtaminer, der er opløselige i et smøremiddel, til passivering af hele eller en del af overfladen af metaldele i en motor, fortrinsvis en skibsmotor, og for at forhindre og/eller reducere metaltab på nævnte metaldele, hvor den fedtaminen er valgt fra:

- Forbindelserne med formel (I):

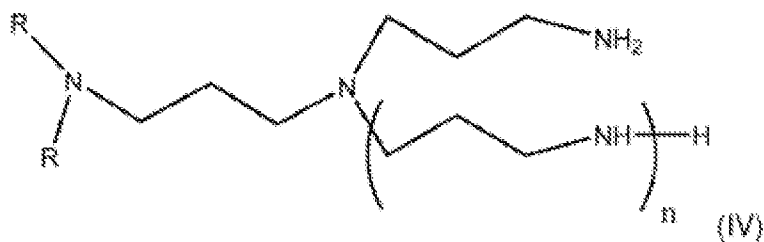


hvor

- R_1 repræsenterer en mættet eller umættet carbonhydridgruppe, lineær eller forgrenet, bestående af fra 12 til 22 carbonatomer,
- R_2 , R_4 eller R_5 repræsenterer uafhængigt et hydrogenatom, en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende mellem 1 og 22 carbonatomer eller en gruppe $(R_6-O)_q-H$, i hvilken R_6 er en mættet, lineær eller forgrenet alkylgruppe, omfattende mellem 2 og 6 carbonatomer, og q repræsenterer et heltal større end eller lig med 1,
- R_3 repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende mellem 2 og 6 carbonatomer,
- m er et heltal mellem 1 og 10, mere fortrinsvis mellem 1 og 6, endnu mere fortrinsvis valgt fra 1, 2 eller 3; eller
- en blanding af en eller flere polyalkylaminer med formel (III) og/eller (IV):



(III),



hvor

- R, som er identisk eller forskellig, repræsenterer en lineær eller forgrenet alkylgruppe omfattende fra 8 til 22 carbonatomer,
- n og z uafhængigt af hinanden repræsenterer 0, 1, 2 eller 3, og
- når z er større end 0, o og p uafhængigt af hinanden repræsenterer 0, 1, 2 eller 3,

nævnte blanding omfattende mindst 3 vægtprocent forgrenede forbindelser, således at mindst én af n eller z er større end eller lig med 1, eller derivater deraf; eller

- en blanding af fedtaminer med formel (I), (III) og/eller (IV).

2. Anvendelse ifølge krav 1, hvori:

- R₁ repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende fra 14 til 22 carbonatomer, og/eller
- R₂, R₄ eller R₅ repræsenterer uafhængigt et hydrogenatom, en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende mellem 14 og 22 carbonatomer, fortrinsvis mellem 16 og 22 carbonatomer; eller en gruppe (R₆-O)_q-H, i hvilken R₆ er en mættet, lineær eller forgrenet alkylgruppe, omfattende mellem 2 og 4 carbonatomer, og q repræsenterer et heltal mellem 1 og 6, fortrinsvis mellem 1 og 4, og/eller
- R₃ repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende mellem 2 og 4 carbonatomer.

3. Anvendelse ifølge krav 1 eller 2, hvori:

- m er lig med 1, 2 eller 3,
- R₁ repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende fra 12 til 20 carbonatomer, fortrinsvis fra 14 til 20 carbonatomer,
- R₂ repræsenterer uafhængigt et hydrogenatom eller en mættet, lineær eller forgrenet alkylgruppe, omfattende fra 1 til 20 carbonatomer,

fortrinsvis fra 16 til 20 carbonatomer, mere fortrinsvis fra 16 til 18 carbonatomer,

- R_3 repræsenterer en mættet og lineær alkylgruppe, omfattende mellem 2 og 6 carbonatomer, fortrinsvis mellem 2 og 4 carbonatomer,
- R_4 og R_5 repræsenterer et hydrogenatom eller en methylgruppe, fortrinsvis et hydrogenatom.

4. Anvendelse ifølge krav 3, hvori:

- m er lig med 3,
- R_1 repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende fra 12 til 20 carbonatomer, fortrinsvis fra 14 til 20 carbonatomer, mere fortrinsvis fra 16 til 20 carbonatomer,
- R_2 repræsenterer uafhængigt et hydrogenatom eller en mættet, lineær eller forgrenet alkylgruppe, omfattende fra 16 til 18 carbonatomer,
- R_3 repræsenterer en ethyl- eller propylgruppe,
- R_4 og R_5 repræsenterer et hydrogenatom.

5. Anvendelse ifølge krav 1 eller 2, hvori:

- m er lig med 1, 2 eller 3,
- R_1 repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende fra 14 til 20 carbonatomer, fortrinsvis fra 16 til 20 carbonatomer,
- R_2 , R_4 og R_5 repræsenterer uafhængigt et hydrogenatom eller en gruppe $(R_6-O)_q-H$, i hvilken R_6 er en mættet, lineær alkylgruppe omfattende mellem 2 og 6 carbonatomer, mere fortrinsvis mellem 2 og 4 carbonatomer, og q repræsenterer et heltal mellem 1 og 6, mere fortrinsvis mellem 1 og 4,
- R_3 repræsenterer en mættet og lineær alkylgruppe, omfattende mellem 2 og 6 carbonatomer, fortrinsvis mellem 2 og 4 carbonatomer.

6. Anvendelse ifølge krav 5, hvori:

- m er lig med 3,
- R_1 repræsenterer en mættet eller umættet alkylgruppe, lineær eller forgrenet, omfattende fra 14 til 20 carbonatomer, fortrinsvis fra 16 til 20 carbonatomer,

- R_2 , R_4 og R_5 repræsenterer uafhængigt et hydrogenatom eller en gruppe $(R_6-O)_q-H$, i hvilken R_6 er en mættet, lineær alkylgruppe omfattende mellem 2 og 4 carbonatomer, og q repræsenterer et heltal mellem 1 og 4,
 - R_3 repræsenterer en ethyl- eller propylgruppe.
7. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af polyalkylaminer med formel (III) og/eller (IV) omfatter mindst 5 vægtprocent af forbindelser med en ren lineær struktur.
8. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af polyalkylaminer med formel (III) og/eller (IV) omfatter mindst 4 vægtprocent, fortrinsvis mindst 5 vægtprocent, fortrinsvis mindst 6 vægtprocent, fortrinsvis mere end 7 vægtprocent, fortrinsvis mere end 7,5 vægtprocent, fortrinsvis mere end 10 vægtprocent, fortrinsvis mere end 20 vægtprocent forgrenede forbindelser, således at mindst n eller z er større end eller lig med 1.
9. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af polyalkylaminer med formel (III) og/eller (IV) mindst omfatter polyalkylaminer med formel (III) og/eller (IV) således, at når n , o , p og z ikke er lig med 0, er de lig med 1 eller 2, fortrinsvis er de lig med 1.
10. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af polyalkylaminer med formel (III) og/eller (IV) mindst omfatter polyalkylaminer med formel (III) og/eller (IV), for hvilke n , o , p eller z uafhængigt repræsenterer 0, 1 eller 2, fortrinsvis repræsenterer de uafhængigt 0 eller 1.
11. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af polyalkylaminer med formel (III) og/eller (IV) mindst omfatter polyalkylaminer med formel (III) og/eller (IV) og deres derivater, for hvilke n , o , p eller z uafhængigt repræsenterer 0, 1 eller 2, fortrinsvis repræsenterer de uafhængigt 0 eller 1.
12. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor blandingen af fedtaminer med formel (I), (III) og/eller (IV) udgør fra 0,1 til 15 vægtprocent, fortrinsvis fra 0,5 til 10 vægtprocent, fortrinsvis fra 0,5 til 8 vægtprocent eller fra 3 til 10 vægtprocent i forhold til den samlede vægt af smøremidlet.

13. Anvendelse ifølge et hvilket som helst af de foregående krav til at forhindre og/eller reducere metaltab på dele i en skibs-, totakts- eller firetaktsmotor under forbrænding af enhver type brændselsolie.
14. Anvendelse ifølge et hvilket som helst af de foregående krav til at forhindre og/eller reducere metaltab på dele i de varme dele, især SPC-zonen, i en skibs-, totakts- eller firetaktsmotor under forbrænding af enhver type brændselsolie.
15. Anvendelse ifølge et hvilket som helst af de foregående krav, hvor brændselsolien har et svovlindhold på under 3,5 vægtprocent i forhold til brændselsoliens samlede vægt.