

(19)



(11)

**EP 2 794 828 B1**

(12)

## EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention  
of the grant of the patent:  
**04.09.2019 Bulletin 2019/36**

(51) Int Cl.:  
**C10M 169/06** <sup>(2006.01)</sup> **C10N 30/06** <sup>(2006.01)</sup>  
**C10N 50/10** <sup>(2006.01)</sup>

(21) Application number: **12808414.2**

(86) International application number:  
**PCT/EP2012/076851**

(22) Date of filing: **21.12.2012**

(87) International publication number:  
**WO 2013/093104 (27.06.2013 Gazette 2013/26)**

### (54) GREASE COMPOSITION

SCHMIERFETTZUSAMMENSETZUNG

COMPOSITION DE GRAISSE

(84) Designated Contracting States:  
**AL AT BE BG CH CY CZ DE DK EE ES FI FR GB  
GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO  
PL PT RO RS SE SI SK SM TR**

(30) Priority: **22.12.2011 JP 2011281987**

(43) Date of publication of application:  
**29.10.2014 Bulletin 2014/44**

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**EP 2 794 828 B1**

**Description**Technical Field of the Invention

5     **[0001]** The present invention relates to a grease composition and, more particularly, to a grease composition with high wear resistance.

Background of the Invention

10    **[0002]** The sliding parts and the rotating parts of machines require some kind of lubricating substrate and lubricating oils or greases are mostly employed therein with, in particular, very many machines utilizing a grease lubrication system where the sealing structure can be simplified and equipment made small and compact. The range of use is extremely broad, encompassing, for example, the various kinds of ball-and-roller bearings and sliding bearings which support rotating bodies, and also the sliding parts of ball joints and links or chains, gear wheels, wires and booms of cranes,  
 15    etc., and the quality demanded of these rises year by year, with the prolongation of machine life and maintenance-free operation being problems common to all machinery. Moreover, in recent years, along with the advances made in machine technology, materials technology and machining precision, fatigue failure and damage to materials has become very low, so the machine life has come to be greatly influenced by the performance of the lubricating oil or grease. Thus, enhancing the lubricating properties of the grease, and solving the problems associated therewith, can contribute greatly  
 20    to the raising of the quality and reliability of machines, and so are extremely important.

**[0003]** Specifically, the cases where the lubricating performance of a grease in the rotating components or sliding parts of a machine falls, and the machine life runs out, may be broadly divided into two. Firstly, there is the case where the grease is subject to oxidation when used at high temperature, so that there is solidification of the grease due to evaporation of the oil component or thermal polymerization, or the grease structure breaks down along with the formation  
 25    of organic acids or aldehydes, resulting in lubrication failure. Secondly, there is the case where, as a result of considerable sliding friction at the machine sliding faces during use at comparatively low speeds under high loads, or at high speeds, a boundary lubrication state tends to come about and the lubricating film of grease becomes extremely thin, so that frequent metal/metal contact occurs and wear increases, leading to damage such as metal separation or seizure, etc.

**[0004]** Measures for overcoming the situation described in the first case above include methods aimed at extending the machine life by effectively incorporating a suitable antioxidant into the grease (see Japanese Patent 2085136), or using, as a grease ingredient, a thickener or a base oil which undergoes little structural or chemical change due to heat, thereby enhancing the function of the grease as a whole so that the machine life is prolonged. Urea-based greases are one example of thermally stable thickeners and recently there have been many technical developments employing such thickeners (see Japanese Patent 4769456). Since they have a higher dropping point than a grease employing a lithium  
 35    soap as a thickener, and since they are outstanding in their heat stability and also in their wear resistance and lubricity, urea greases have come to be used in a wide range of fields. For example, in the automotive industry, the levels demanded for heat resistance, wear resistance and friction characteristics in regard to various kinds of motor vehicle components such as CVJs (constant velocity joints), electric power steering, alternator bearings and wheel bearings, etc., have been steadily increasing, so there are many cases where the outstanding performance and additive technology  
 40    of the urea greases are being applied.

**[0005]** Moreover, since motor vehicles are used all around the world, the grease employed in motor vehicle components is designed and produced taking into account regular use under extremely cold conditions of about - 40°C and also at high temperatures of 100°C and above (radiant heat from the engine compartment + heat emitted from the road surface) so, as well as obtaining low stabilized torque characteristics over a wide temperature range from low to high temperatures,  
 45    a grease is demanded which does not show deficient oiling due to a drop in viscosity at high temperatures and which also itself has a long life to match the life of the vehicle.

**[0006]** For enhancing the low temperature characteristics, a technique has been disclosed in which a methacrylate polymer is incorporated into a synthetic hydrocarbon oil (poly- $\alpha$ -olefin), ester type base oil or glycol type base oil having outstanding low temperature fluidity as a grease base oil, but further improvements in quality and performance are  
 50    demanded (see JP-A-2006-77119).

**[0007]** Measures for overcoming the situation described in the second case above include methods where the grease base oil viscosity is raised and polymer or other viscosity enhancing agent incorporated, so that the lubricating film is thickened and metal/metal contact suppressed (see JP-A-2008-69282), and methods where an antiwear agent, an extreme pressure agent, a solid lubricant or other load-resisting additive is incorporated into the grease so that, by the  
 55    chemical or physical action of this additive, a coating or solid film is formed between the sliding faces and the surfaces thereby protected (see Japanese Patent 3833756 and JP-A-2-18497). However, depending on the type of additive, the structure of the grease may be altered, and there are also many additives which have an adverse effect on the machine components. For example, thiophosphates of high acid value have the disadvantage of readily reacting with the free

alkali of a soap-based grease, and olefin sulphide type additives may bring about hardening of a urea grease with the passage of time. Furthermore, in the case of additives with strong chemical activity, there are many issues such as discoloration or the corrosion of metals being caused and the strength of sealing materials like nitrile, acrylic or urethane rubbers being lowered.

**[0008]** US 3 130 161 A discloses a lubricating oil and anti-wear additives therefor.

**[0009]** Consequently, in terms of grease lubrication, being able to maintain a suitable oil film and form an adsorbed film at the lubricated sliding faces, without having any adverse effects on the grease structure, is extremely effective in terms of stabilizing the quality of motor vehicle parts and industrial components, and also in ensuring reliability and prolonging the realized machine life.

#### Summary of the Invention

**[0010]** The present invention addresses the problem of providing a grease composition with high wear resistance. As a result of painstaking research against this background, the present inventors have discovered that it is possible to raise the wear resistance of a grease by employing a poly(meth)acrylate derivative which contains hydroxyl groups and, in this way, it is possible to solve the aforesaid problem.

**[0011]** Accordingly the present invention provides a grease composition which is characterized in that, in a grease composition containing a base oil and a thickener, there is employed, as an additive, a poly(meth)acrylate derivative containing hydroxyl groups.

**[0012]** The grease composition relating to the present invention displays the effect of higher wear resistance when compared to conventional grease compositions.

#### Detailed Description of the Invention

**[0013]** The grease composition of this embodiment contains, as indispensable constituent components, "base oil", "thickener" and "additive". Below, the individual components contained in the grease composition, the amounts (blended quantities) of the components in the grease composition, the method of producing the grease composition, the properties of the grease composition, and applications of the grease composition are explained in turn.

**[0014]** The base oil employed in the grease composition of this mode of the invention is not particularly restricted. For example, there can be appropriately employed the mineral oils, synthetic oils, animal/vegetable oils, and mixtures thereof, used in normal grease compositions. As specific examples, the base oils belonging to Group I, Group II, Group III and Group IV, etc., of the API (American Petroleum Institute) base oil categories can be used on their own or in the form of mixtures.

**[0015]** The Group I base oils include paraffinic mineral oils obtained by, for example, applying a suitable combination of purification means such as solvent refining, hydro-refining and dewaxing, etc., to the lubricating oil fraction obtained by the atmospheric distillation of crude oil. Group II base oils include paraffinic mineral oils obtained by, for example, applying a suitable combination of purification means such as hydro-cracking and dewaxing, etc., to the lubricating oil fraction obtained by the atmospheric distillation of crude oil. The Group II base oils produced by a hydro-refining method such as the Gulf Co. method have a total sulphur component content of less than 10 ppm and an aromatics content of no more than 5%, and can be favourably employed in the present invention. Group III base oils and Group II+ base oils include, for example, the paraffinic mineral oils produced by subjecting the lubricating oil fraction obtained by the atmospheric distillation of crude oil to high level hydro-refining, and also base oils refined by the Isodewaxing process where the wax formed in the dewaxing process is converted to isoparaffins, as well as the base oils produced by the Mobil Wax Isomerization process, and these too can be favourably used in this mode of the invention.

**[0016]** Examples of the synthetic oils include polyolefins, the diesters of dibasic acids like dioctyl sebacate, polyol esters, alkylbenzenes, alkylnaphthalenes, esters, polyoxyalkylene glycols, polyoxyalkylene glycol esters, polyoxyalkylene glycol ethers, polyphenyl ethers, dialkyl diphenyl ethers, fluorine-containing compounds (perfluoropolyethers, fluorinated polyolefins and the like), and silicones, etc. The aforesaid polyolefins include various types of olefin polymer and also their hydrogenated products. Any olefin may be used, and examples are ethylene, propylene, butene, and  $\alpha$ -olefins with five or more carbon atoms. In the production of the polyolefin, a single type of aforesaid olefin may be used on its own, or a combination of two or more types may be employed. In particular, from the point of view of enhancing the low temperature fluidity and the low temperature lubricity, the polyolefins referred to as poly- $\alpha$ -olefin (PAO) synthetic oils are preferred, and these are Group IV base oils. Since the viscosity index of poly- $\alpha$ -olefin synthetic oils is high, there is little lowering in viscosity at high temperatures and their capacity to maintain an oil film is high, in addition to which the viscosity is not excessively raised at low temperatures and suitable fluidity is maintained, so there is little fall in lubricity. When this outstanding low temperature behaviour is used for the base oil of a grease, it acts extremely effectively. For example, if a lubricating oil which has extremely high viscosity at low temperature, and so loses fluidity, is used as the grease base oil, the fluidity of the grease is totally impaired and there is practically no supply of lubricating oil to the

sliding faces of the machine components, so that wear can progress, whereas, with a grease in which there is incorporated a base oil having outstanding low temperature fluidity, such as a poly- $\alpha$ -olefin synthetic oil, there is no loss in fluidity and the lubricating oil can be appropriately supplied to the sliding faces, so that the lubricating function is maintained and wear suppressed.

**[0017]** The oils obtained by a GTL (gas to liquid) process, which are oils synthesized from natural gas in a liquid fuel conversion technique employing the Fischer Tropsch method, have an extremely low sulphur component and aromatic component content when compared to the mineral oil base oils obtained by the refining of crude oil, and the paraffin structure proportion is extremely high, with the result that they have outstanding oxidation stability and an extremely low evaporation loss, so these can be favourably employed as the base oil in the present mode of the invention.

**[0018]** The thickener used as an indispensable component in this mode of the invention are the compounds represented by general formula (I) :



(where  $R^1$  and  $R^3$  each represents a  $C_{8-12}$  aliphatic hydrocarbon group, and  $R^2$  represents a  $C_{6-15}$  divalent aromatic group). Here, it is preferred that  $R^1$  and  $R^3$  are mutually independently an eight-carbon octyl group or a twelve-carbon lauryl group. Furthermore,  $R^2$  is preferably the diphenylmethane group. In more preferred embodiments, there are used the compounds where  $R^2$  is the diphenylmethane group and (1)  $R^1$  and  $R^3$  are eight-carbon octyl groups, or (2-1) a mixture of the compound where  $R^1$  and  $R^3$  are each eight-carbon octyl groups and the compound where  $R^1$  and  $R^3$  are twelve-carbon lauryl groups, (2-2) the compound where either  $R^1$  or  $R^3$  is the eight-carbon octyl group while the other from amongst  $R^1$  and  $R^3$  is the twelve-carbon lauryl group, and (2-3) a mixture of one or both of the compounds in (2-1) and the compound in (2-2). Of these, (2-1) to (2-3) are particularly preferred.

**[0019]** In the grease composition in this mode of the invention, it is possible to jointly employ another thickener along with the main thickener {which is, for example, an aforesaid urea compound (such as an alkyl diurea)}. Thus, in the case where there is used a urea compound as the main thickener, it is possible to employ, as this other thickener, tribasic calcium phosphate, an alkali metal soap, an alkali metal complex soap, an alkaline earth metal soap, an alkaline earth complex soap, an alkali metal sulphonate, an alkaline earth metal sulphonate, other metal soaps, a metal terephthalamate salt, clay, a silica aerogel or other type of silica (silicon oxide), or a fluoropolymer such as polytetrafluoroethylene, etc., and one of these can be used on its own or two or more can be used in combination. As well as these, it is also possible to employ any material which can confer a viscosity raising effect.

**[0020]** The additive employed as an indispensable component in this embodiment is a poly(meth)acrylate which contains hydroxyl groups. This poly(meth)acrylate containing hydroxyl groups is a copolymer, and comprises a copolymer based on an alkyl (meth)acrylate having a  $C_{1-20}$  alkyl group, and a hydroxyl-group-containing vinyl monomer, as the indispensable constituent monomers thereof.

**[0021]** Specific examples of the aforesaid alkyl (meth) acrylate (a) with a  $C_{1-20}$  alkyl group are

(a1) alkyl (meth) acrylates with a  $C_{1-4}$  alkyl group: examples of which are methyl (meth)acrylate, ethyl (meth)acrylate, n- or iso-propyl (meth)acrylate, and n-, iso- or sec-butyl (meth)acrylate,

(a2) alkyl (meth) acrylates with a  $C_{8-20}$  alkyl group:

examples of which are n-octyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, n-decyl (meth)acrylate, n-isodecyl (meth)acrylate, n-undecyl (meth)acrylate, n-dodecyl (meth)acrylate, 2-methylundecyl (meth)acrylate, n-tridecyl (meth)acrylate, 2-methyldodecyl (meth)acrylate, n-tetradecyl (meth)acrylate, 2-methyltridecyl (meth)acrylate, n-pentadecyl (meth)acrylate, 2-methyltetradecyl (meth)acrylate, n-hexadecyl (meth)acrylate, n-octadecyl (meth)acrylate, n-eicosyl (meth)acrylate, n-docosyl (meth)acrylate, Dovanol 23 [a  $C_{12}/C_{13}$  oxo-alcohol mixture produced by the Mitsubishi Chemical Corp.] methacrylate and Dovanol 45 [a  $C_{13}/C_{14}$  oxo-alcohol mixture produced by the Mitsubishi Chemical Corp.] methacrylate, etc.,

(a3) alkyl (meth)acrylates with a  $C_{5-7}$  alkyl group:

examples of which are n-pentyl (meth)acrylate and n-hexyl (meth)acrylate, etc.

**[0022]** Amongst (a1) to (a3) above, the monomers belonging to (a1) and (a2) are preferred, with those belonging to (a2) further preferred. Moreover, in terms of solubility in the base oil, from amongst the monomers in (a2), those with 10 to 20 carbons in the alkyl group are preferred and those with 12 to 14 carbons are further preferred.

**[0023]** The aforesaid hydroxyl-group-containing vinyl monomer (b), from which the copolymer is composed along with the alkyl (meth)acrylate having a  $C_{1-20}$  alkyl group, is a vinyl monomer containing one or more (preferably 1 or 2) hydroxyl groups in the molecule. Specific examples are

(b1) hydroxyalkyl ( $C_{2-6}$ ) (meth) acrylates:

such as 2-hydroxyethyl (meth)acrylate, 2- or 3-hydroxypropyl (meth)acrylate, 2-hydroxybutyl (meth)acrylate, 1-me-

thyl-2-hydroxyethyl (meth)acrylate, etc.,

(b2) mono- or di-hydroxyalkyl(C<sub>1-4</sub>)-substituted (meth)acrylamides:

such as N,N-dihydroxymethyl (meth)acrylamide, N,N-dihydroxypropyl (meth)acrylamide, N,N-di-2-hydroxybutyl (meth)acrylamide, etc.,

(b3) vinyl alcohol (formed by the hydrolysis of vinyl acetate units),

(b4) C<sub>3-12</sub> alkenols:

such as (meth)allyl alcohol, crotyl alcohol, isocrotyl alcohol, 1-octenol, 1-undecenol, etc.,

(b5) C<sub>4-12</sub> alkene diols:

such as 1-buten-3-ol, 2-buten-1-ol, 2-buten-1,4-diol, etc.,

(b6) hydroxyalkyl (C<sub>1-6</sub>) alkenyl (C<sub>3-10</sub>) ethers:

such as 2-hydroxyethyl propenyl ether, etc.,

(b7) hydroxyl-group-containing aromatic monomers:

such as o-, m- or p-hydroxystyrene, etc.,

(b8) polyhydric alcohols (from tri- to octa-hydric):

such as alkane polyols, as well as their intramolecular or intermolecular dehydro-condensates, and sugar (e.g. glycerol, pentaerythritol, sorbitol, sorbitan, diglycerol or sucrose) alkenyl(C<sub>3-10</sub>) ethers or (meth)acrylates (e.g. sucrose (meth)allyl ether), etc.,

(b9) vinyl monomers containing a polyoxyalkylene chain and hydroxyl group(s):

such as the mono(meth)acrylate or mono(meth)allyl ether of a polyoxyalkylene glycol (C<sub>2-4</sub> alkylene group; degree of polymerization 2 to 50) or of a polyoxyalkylene polyol {a polyoxyalkylene ether of an aforesaid tri- to octa-hydric alcohol (C<sub>2-4</sub> alkyl group; degree of polymerization 2 to 100)} {examples of which are polyethylene glycol (degree of polymerization 2-9) mono(meth)acrylate, polypropylene glycol (degree of polymerization 2-12) mono(meth)acrylate, polyethylene glycol (degree of polymerization 2-30) mono(meth)allyl ether}, and the like.

**[0024]** It is also possible to copolymerize other monomers with the aforesaid monomers (a) and (b) in the aforesaid poly(meth)acrylate copolymer containing hydroxyl groups, and, for example, there may be used a monomer (c) which contains a nitrogen atom. Specific examples thereof are

(c1) nitro-group-containing monomers: such as 4-nitrostyrene, etc.,

(c2) vinyl monomers containing primary, secondary or tertiary amino groups: examples of which are

(c2-1) vinyl monomers containing a primary amino group; such as C<sub>3-6</sub> alkenylamines [(meth)allylamine, crotylamine, etc.], aminoalkyl(C<sub>2-6</sub>) (meth)acrylates [aminoethyl (meth)acrylate, etc.],

(c2-2) vinyl monomers containing a secondary amino group; such as alkyl (C<sub>1-6</sub>) aminoalkyl (C<sub>2-6</sub>) (meth)acrylates [tert-butylaminoethyl methacrylate, methylaminoethyl (meth)acrylate, etc.], diphenylamine (meth)acrylamides [4-diphenylamine (meth)acrylamide, 2-diphenylamine (meth)acrylamide, etc.], C<sub>6-12</sub> dialkenylamines [di(meth)allylamine, etc.],

(c2-3) vinyl monomers containing a tertiary amino group; such as dialkyl (C<sub>1-4</sub>) -aminoalkyl (C<sub>2-6</sub>) (meth)acrylates [dimethylaminoethyl (meth)acrylate, diethylaminoethyl (meth)acrylate, etc.], dialkyl(C<sub>1-4</sub>)aminoalkyl (C<sub>2-6</sub>) (meth)acrylamides [dimethylaminoethyl - (meth)acrylamide, diethylaminoethyl(meth)acrylamide, dimethylaminopropyl-(meth)acrylamide, etc.], aromatic vinyl monomers containing a tertiary amino group [N,N-dimethylaminostyrene, etc.],

(c2-4) vinyl monomers containing a nitrogen heterocycle [morpholinoethyl (meth)acrylate, 4-vinylpyridine, 2-vinylpyridine, N-vinylpyrrole, N-vinylpyrrolidone, N-vinylthiopyrrolidone, etc.],

(c3) amphoteric vinyl monomers: examples of which are

N- (meth)acryloyloxy (or amino) alkyl (C<sub>1-10</sub>) N,N-dialkyl (C<sub>1-5</sub>) ammonium-N-alkyl (C<sub>1-5</sub>) carboxylates (or sulphates), like N-(meth)acryloyloxyethyl N,N-dimethylammonium N-methyl carboxylate, N-(meth)acryloylaminopropyl N,N-dimethylammonium N-methyl carboxylate and N-(meth)acryloyloxyethyl N,N-dimethylammonium propyl sulphate, etc.; and

(c4) monomers containing a nitrile group, such as (meth)acrylonitrile, and the like.

**[0025]** Moreover, other examples of such copolymerized monomers are aliphatic hydrocarbon-type vinyl monomers (d). Examples of these are C<sub>2-20</sub> alkenes [ethylene, propylene, butene, isobutylene, pentene, heptene, diisobutylene, octene, dodecene, octadecene, etc.] and C<sub>4-12</sub> alkadienes [butadiene, isoprene, 1,4-pentadiene, 1,6-heptadiene, 1,7-octadiene, etc.].

**[0026]** Again, there are alicyclic hydrocarbon-type vinyl monomers (e): examples of which are cyclohexene, (di)cyclopentadiene, pinene, limonene, indene, vinylcyclohexene and ethylidene bicycloheptene, etc.

**[0027]** Additionally, there are aromatic hydrocarbon-type vinyl monomers (f): examples of which are styrene,  $\alpha$ -methylstyrene, vinyltoluene, 2,4-dimethylstyrene, 4-ethylstyrene, 4-isopropylstyrene, 4-butylstyrene, 4-phenylstyrene, 4-cyclohexylstyrene, 4-benzylstyrene, 4-crotylbenzene and 2-vinylnaphthalene, etc.

**[0028]** Yet again, there are vinyl esters, vinyl ethers and vinyl ketones (g): examples of which are the vinyl esters of saturated  $C_{2-12}$  fatty acids [vinyl acetate, vinyl propionate, vinyl butyrate, vinyl octanoate, etc.],  $C_{1-12}$  alkyl, aryl or alkoxy-alkyl vinyl ethers [methyl vinyl ether, ethyl vinyl ether, propyl vinyl ether, butyl vinyl ether, 2-ethylhexyl vinyl ether, phenyl vinyl ether, vinyl 2-methoxyethyl ether, vinyl 2-butoxyethyl ether, etc.] and  $C_{1-8}$  alkyl or aryl vinyl ketones [methyl vinyl ketone, ethyl vinyl ketone, phenyl vinyl ketone, etc.].

**[0029]** Moreover, there are the esters of unsaturated polycarboxylic acids (h): examples of which are the alkyl, cycloalkyl and aralkyl esters of unsaturated polycarboxylic acids, in particular the  $C_{1-8}$  alkyl diesters of unsaturated dicarboxylic acids [such as maleic acid, fumaric acid and itaconic acid] [like dimethyl maleate, dimethyl fumarate, diethyl maleate and dioctyl maleate, etc.].

**[0030]** It is also possible to use vinyl monomers which contain a polyoxyalkylene chain (but not containing hydroxyl groups) (i): examples of these being the mono-(meth) acrylates of a mono-alkyl ( $C_{1-18}$ ) ether of a polyoxyalkylene glycol ( $C_{2-4}$  alkylene group; degree of polymerization 2-50) or of a polyoxyalkylene polyol [polyoxyalkylene ether of an aforesaid tri- to octa-hydric alcohol ( $C_{2-4}$  alkyl group; degree of polymerization 2-100)], [such as methoxy polyethylene glycol (Mw 110-310) (meth)acrylate, or the (meth)acrylate of a lauryl alcohol ethylene oxide (2-30 mol) adduct, etc.]

**[0031]** Again, as monomer in the aforesaid copolymerization, it is also possible to use carboxyl group-containing vinyl monomers (j): examples of which are vinyl monomers containing a single carboxyl group like unsaturated monocarboxylic acids [(meth)acrylic acid,  $\alpha$ -methyl (meth) acrylic acid, crotonic acid, cinnamic acid, etc.] and the monoalkyl ( $C_{1-8}$ ) esters of unsaturated dicarboxylic acids [maleic acid monoalkyl ester, fumaric acid monoalkyl ester, itaconic acid monoalkyl ester, etc.]; and vinyl monomers containing two or more carboxyl groups like maleic acid, fumaric acid, itaconic acid and citraconic acid, etc.

**[0032]** Of the additional monomers (c), (d), (e), (f), (g), (h), (i) and (j) described above, the monomers (c) are preferred, and two or more of these monomers (c) may be used in combination. Amongst the monomers (c), the monomers (c2) are preferred, with dimethylaminoethyl (meth)acrylate and diethylaminoethyl (meth)acrylate further preferred.

**[0033]** The hydroxyl value (hydroxyl group value) of the poly(meth)acrylate containing hydroxyl groups used as the additive lies between 10 and 100, preferably between 20 and 50, and more preferably between 25 and 35. The hydroxyl value is a numerical value obtained by measurement based on JIS K3342 (1961), and it expresses the amount of hydroxyl groups in the additive.

**[0034]** When this hydroxyl-group-containing poly(meth)acrylate is incorporated into the grease, there are no adverse effects on the grease structure, a suitable oil film is secured at the lubricated sliding faces and, furthermore, the chemical structure containing hydroxyl groups acts effectively and an adsorbed film is formed at the lubricated sliding faces. Consequently, with a grease in which this hydroxyl-group-containing poly(meth)acrylate has been incorporated, it is possible to provide ideal lubricity at the sliding faces and suppress wear to the lubricated parts of motor vehicle components and industrial components, and it is extremely effective in achieving stability and reliability in terms of the quality of machine parts and extending the life thereof. A major difference between a lubricating oil and a grease is that, in the case of a grease, the thickener and additive forming the framework of the grease structure interact together and so, whereas an effect may be manifested in the case of a lubricating oil, with a grease there may be no effect at all, or, alternatively, wear may actually be promoted. Consequently, an additive which is stable and confers effective lubricity in terms of the grease is extremely favourable, as in the case of this present technique.

**[0035]** The grease composition of this mode of the invention may also optionally contain further additives such as antioxidants, rust inhibitors, oiliness agents, extreme pressure agents, anti-wear agents, solid lubricants, metal deactivators, polymers, metal-based cleaning agents, non metal-based cleaning agents, colouring agents, and the like. Examples of the antioxidants include 2,6-di-*tert*-butyl-4-methylphenol, 2,6-di-*tert*-butyl-p-cresol, p,p'-dioctyldiphenyl-amine, N-phenyl- $\alpha$ -naphthylamine and phenothiazine. Examples of the rust inhibitors include oxidized paraffin, carboxylic acid metal salts, sulphonic acid metal salts, carboxylic acid esters, sulphonic acid esters, salicylic acid esters, succinic acid esters, sorbitan esters and various kinds of amine salts. Examples of the oiliness agents, extreme pressure agents and anti-wear agents include zinc dialkyldithiophosphate sulphide, zinc diallyldithiophosphate sulphide, zinc dialkyldithiocarbamate sulphide, zinc diallyldithiocarbamate sulphide, molybdenum dialkyldithiophosphate sulphide, molybdenum diallyldithiophosphate sulphide, molybdenum dialkyldithiocarbamate sulphide, organomolybdenum complexes, olefin sulphide, triphenyl phosphate, triphenyl-phosphorothionate, tricresyl phosphate and other phosphate esters, and sulphurized oil/fats, etc. Examples of the solid lubricants include molybdenum disulphide, graphite, boron nitride, melamine cyanurate, PTFE (polytetrafluoroethylene), tungsten disulphide, graphite fluoride and calcium phosphate. Examples of the metal deactivators include N,N'-disalicylidene-1,2-diaminopropane, benzotriazole, benzimidazole, benzothiazole and thiadiazole, etc. Examples of the polymers include polybutene, polyisobutene, polyisobutylene, polyisoprene, polymethacrylate, and the like. Examples of the metal-based cleaning agents are metal sulphonates, metal salicylates and metal phenates. Examples of the non metal-based cleaning

agents include succinimides and the like. However, these examples are not intended to restrict the invention.

**[0036]** Next, explanation is provided of the amounts of the respective components employed in the grease composition relating to this mode of the invention. Unless stated otherwise, % refers to percentage by mass.

**[0037]** The blended amount of base oil is preferably from 60-95 mass%, more preferably 70-90 mass% and still more preferably 75-90 mass%, taking the total grease composition as 100 mass%.

**[0038]** The amount of thickener incorporated is from 1-20 mass% , preferably 2-17 mass% and more preferably 3-15 mass%, taking the total grease composition as 100 mass%.

**[0039]** The amount of hydroxyl-group-containing poly(meth)acrylate incorporated is preferably from 2-20 mass% and more preferably 4-10 mass%, taking the total grease composition as 100 mass%.

**[0040]** The amount of the other additives is, for example, from 0.03 to 20 mass% by total of these optional components, taking the total grease composition as 100 mass%.

**[0041]** A particularly preferred blending embodiment (in particular in regard to the thickener and the PMA containing hydroxyl groups) is, taking the case where the type of thickener is from amongst (2-1) to (2-3) above, a thickener content = 7.5-15 mass% (more preferably 10-15 mass%), and a hydroxy-group-containing PMA content = 7.5-15 wt% in the case where the type of this PMA containing hydroxyl groups is one where (hydroxyl value = 20-50, weight average molecular weight =  $1.0 \times 10^4$  to  $2.0 \times 10^4$ ) .

**[0042]** The grease composition of this mode of the invention can be produced by generally-used grease production methods and is not particularly restricted but, as an example of the method for producing a urea grease composition, a diurea thickener obtained by the reaction of 1 mol of diisocyanate and 2 mol of primary monoamine as the starting materials for the urea thickener, or a tetraurea grease thickener obtained by the reaction of 2 mol of diisocyanate and 1 mol of primary diamine plus 2 mol of primary monoamine as the starting materials for the urea thickener, or a triurea-monourethane obtained by the reaction of 2 mol of diisocyanate, 1 mol of primary diamine, 1 mol of primary monoamine plus 1 mol of mono-alcohol as the starting materials for the urea thickener, is synthesized along with the base oil in each case in a grease production kettle, to produce the grease used. With regard to a more specific production method, having carried out the synthesis reaction of the particular thickener in the base oil, the temperature is raised to a temperature of about 180°C, after which cooling is performed and, at a temperature of 100-80°C, the additive {the hydroxyl-group-containing poly(meth)acrylate derivative} is incorporated and thorough mixing performed, following which the mixture is cooled to room temperature. Thereafter, a homogenous grease composition can be obtained using a kneading machine (such as a triple roll mill or the like).

**[0043]** The dropping point of the grease composition in this mode of the invention is preferably at least 180°C, more preferably at least 210°C, still more preferably at least 250°C and in particular over or above 260°C. If the dropping point of the grease composition is at least 180°C, it is thought that the possibility of lubrication problems arising such as, for example, loss in viscosity at high temperature and accompanying leakage or seizing, etc., is suppressed. The dropping point refers to the temperature at which there is breakdown of the thickener structure in the case of a grease possessing viscosity, when the temperature is raised. Measurement of the dropping point can be carried out in accordance with JIS K2220 8.

**[0044]** In a consistency test, the consistency of the grease in this mode of the invention preferably lies between grade 000 and grade 6 (85-475), more preferably between grade 0 and grade 4 (175-385), and still more preferably between grade 1 and grade 3 (220-340). The consistency denotes the physical hardness of the grease. In measuring the consistency, measurement of the worked penetration may be carried out in accordance with JIS K2220 7.

**[0045]** Friction testing was carried out based on the high speed four ball wear test in ASTM D2596, using steel ball bearings under conditions comprising a rotation rate of 1200 rpm, a load of 40 kgf and ambient temperature (no temperature control) for 30 minutes, and then evaluation conducted from the wear-scar diameter on the stationary balls.

**[0046]** Not only can the grease composition of this mode of the invention be employed for generally-used machinery, bearings and gears, etc., but it can also show outstanding performance under more severe conditions, such as under high temperature conditions. For example, in the case of motor vehicles, it can be employed favourably in the lubrication of the starter, the alternator, various types of actuator unit and other engine peripherals, the driveshaft, constant velocity joints (CVJs), wheel bearings, the clutch and other regions of the powertrain, the electric power steering (EPS), braking equipment, ball joints, door hinges, handles, the cooling fan motor, brake expanders and various other types of components, etc. Furthermore, it is suitable for the various lubricated regions of power shovels, bulldozers, cranes and other types of construction machinery, and in the railway industry or paper industry, or for forestry equipment, agricultural machinery, chemical plant, wind turbines, power generators, drying ovens, copiers, rolling stock, and the screw joints of seamless pipes, etc., in particular sites subject to high temperatures/high loads. Other suggested applications include hard disk bearings, plastic lubrication, cartridge grease and the like, and satisfactory adaptation to such applications too is possible.

Examples

**[0047]** Next, the present invention is explained in still more detail by means of working examples and comparative examples, but the invention is not to be restricted in any way by the examples provided.

**[0048]** The starting materials employed in the working examples and comparative examples were as follows.

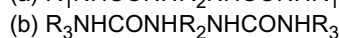
Starting Materials for the Thickeners

**[0049]** With regard to thickeners A and B, diurea components with the following chemical structures were employed as thickeners.

Thickener A: urea type I



Thickener B: urea type II



Thickener C: urea type III



(In these formulae,  $R_2$  is the diphenylmethane group and  $R_1$  is the eight-carbon octyl group,  $R_3$  is the twelve-carbon lauryl group and  $R_4$  is the six-carbon phenyl group.)

Thickener D: This was a commercially-available lithium 12-hydroxystearate for industrial use.

Base Oils A to C**[0050]**

Base oil A: This is a paraffinic mineral oil obtained by dewaxing/solvent refining, and belongs to Group I; its kinematic viscosity is 14.28 mm<sup>2</sup>/s at 100°C and 144.9 mm<sup>2</sup>/s at 40°C; and its viscosity index is 96.

Base oil B: This is a poly- $\alpha$ -olefin (PAO) synthetic oil belonging to Group IV, and it comprises a mixture of a low viscosity PAO and a high viscosity PAO so as to adjust the kinematic viscosity at 100°C to 15.4 mm<sup>2</sup>/s. The kinematic viscosity of the PAO mixture at 40°C is 118.9 mm<sup>2</sup>/s, and its viscosity index is 136.

Base oil C: This is an oil obtained by means of a GTL (gas to liquid) process, synthesized by the Fischer Tropsch method, and belongs to Group III; its kinematic viscosity is 8.2 mm<sup>2</sup>/s at 100°C and 47.9 mm<sup>2</sup>/s at 40°C; and its viscosity index is 144.

Additives**[0051]**

Additive A: Hydroxyl-group-containing PMA (commercial name: Aclube A-1070, produced by Sanyo Chemical Industries Ltd; Mw =  $1.7 \times 10^4$ , hydroxyl value 30)

Additive B: Hydroxyl-group-free PMA (commercial name: Aclube V-815, produced by Sanyo Chemical Industries Ltd; Mw =  $2.1 \times 10^4$ )

Additive C: Hydroxyl-group-free PMA (commercial name: Aclube 812, produced by Sanyo Chemical Industries Ltd; Mw =  $3.3 \times 10^4$ )

Additive D: Hydroxyl-group-free PMA (commercial name: Aclube C728, produced by Sanyo Chemical Industries Ltd; Mw =  $4.5 \times 10^4$ )

Additive E: Ethylene- $\alpha$ -olefin copolymer (commercial name: Lucant HC100, produced by Mitsui Chemicals Inc.)

Additive F: Polybutene (commercial name: Nisseki Polybutene HV-100, produced by the JX Nippon Oil & Energy Corp.)

Working Example 1

**[0052]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 1 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared Group I base oil, i.e. base oil A, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine and laurylamine were respectively dissolved in the remaining 40 wt% of the Group I base oil, and then introduced, in turn, into the trial-production kettle, to bring about reaction and formation of the diurea type II thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 1 was obtained using a triple roll mill.

Working Example 2

**[0053]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 2 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared synthetic oil, i.e. base oil B, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine was dissolved in the remaining 40 wt% of the synthetic oil, and introduced into the trial-production kettle, to bring about reaction and formation of the diurea type I thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 2 was obtained using a triple roll mill.

Working Example 3

**[0054]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 3 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared Group III base oil, i.e. base oil C, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine was dissolved in the remaining 40 wt% of the Group III base oil, and introduced into the trial-production kettle, to bring about reaction and formation of the diurea type I thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 3 was obtained using a triple roll mill.

Working Examples 4-6

**[0055]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the greases according to Working Examples 4 to 6 respectively obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for each of the diurea grease thickeners used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared Group I base oil, i.e. base oil A, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine and laurylamine were respectively dissolved in the remaining 40 wt% of the Group I base oil and introduced, in turn, into the trial-production kettle, to bring

about reaction and formation of the diurea type II thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous greases of Working Examples 4 to 6 were each obtained using a triple roll mill. In Working Examples 4 to 6 the amount of base oil was balanced in accordance with the differing amounts of additive used, and again the amounts of thickener too were slightly adjusted.

#### Working Example 7

**[0056]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 7 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared Group I base oil, i.e. base oil A, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine was dissolved in the remaining 40 wt% of the Group I base oil, and introduced into the trial-production kettle, to bring about reaction and formation of the diurea type I thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 7 was obtained using a triple roll mill.

#### Working Example 8

**[0057]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 8 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, the total amount of the previously-prepared Group I base oil, i.e. base oil A, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, the octylamine was mixed with the synthetic oil, i.e. base oil B, and the laurylamine was mixed and dissolved in base oil C belonging to Group III, and these introduced in turn into the trial-production kettle, to bring about reaction and formation of the diurea type II thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 8 was obtained using a triple roll mill.

#### Working Example 9

**[0058]** The starting materials, base oil and additive were prepared and measured out in the proportions shown in Table 1, and the grease according to Working Example 9 obtained by the usual method for producing a urea grease. The mol ratio of the diisocyanate and primary amine which constituted the starting materials for the diurea grease thickener used here was a ratio of 1 : 2. In terms of the detailed production method, 60 wt% of the total amount of the previously-prepared Group I base oil, i.e. base oil A, was introduced into a sealed grease trial-production kettle, immediately after which the diphenylmethane-4,4'-diisocyanate starting material for the urea thickener was added and, while stirring, the temperature raised to 50°C. Next, aniline was mixed and dissolved in the remaining 40 wt% of the Group I base oil, and introduced into the trial-production kettle, to bring about reaction and formation of the diurea type III thickener. Heating was then continued and the temperature raised to about 180°C, to bring about stabilization of the thickener structure. Subsequently, cooling was commenced and, at a temperature of 80°C during the cooling process, additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was added, and, after thorough stirring and mixing, cooling was continued down to room temperature. Thereafter, the homogeneous grease of Working Example 9 was obtained using a triple roll mill.

Working Example 10

**[0059]** Commercially-available lithium 12-hydroxystearate for industrial use and the Group I base oil, i.e. base oil A, were prepared in the proportions shown in Table 1, and the entire amount of this base oil and lithium 12-hydroxystearate was introduced into a sealed grease trial-production kettle, and the temperature raised to 230°C while stirring. After confirming that the lithium 12-hydroxystearate had completely dissolved, the temperature was gradually reduced to 80°C. Additive A, that is to say the hydroxyl-group-containing poly(meth)acrylate derivative, was then added at this temperature of 80°C and, after thorough stirring and mixing, cooling continued down to room temperature. Thereafter, the homogeneous grease of Working Example 10 was obtained using a triple roll mill.

Comparative Example 1 to Comparative Example 3

**[0060]** Using the production method described in Working Example 6, thickener formation reactions were carried out and comparative greases respectively obtained, where Comparative Example 1 was the base grease in which no additive was incorporated. In Comparative Example 2, additive B was incorporated. In Comparative Example 3, additive C was incorporated.

Comparative Example 4 to Comparative Example 6

**[0061]** Using the production method described in Working Example 1, thickener formation reactions were carried out and comparative greases respectively obtained, with additive D being incorporated in Comparative Example 4. In Comparative Example 5, additive E was incorporated. In Comparative Example 6, additive F was incorporated.

**[0062]** The results obtained are shown in Table 1. It can be seen from Table 1 that, with the greases relating to Working Examples 1 to 10, excellent wear resisting performance is obtained in each case (wear-scar diameter 0.39-0.52).

**[0063]** Working examples 9-10 are for reference.

Table 1

	Working Example										Comparative Example					
	1	2	3	4	5	6	7	8	9	10	1	2	3	4	5	6
Thickener A (mass%)		12.5	12.5				6.5				13.5	12.5	12.5			
Thickener B (mass%)	12.5			11.5	13	13.5		3.5						12.5	12.5	12.5
Thickener C (mass%)									18.5							
Thickener D (mass%)										9.5						
Base Oil A (mass%)	77.5	-	-	68.5	82.0	83.5	83.5	28.5	71.5	80.5	86.5	77.5	77.5	77.5	77.5	77.5
Base Oil B (mass%)	-	77.5	-	-	-	-	-	29.0			-	-	-	-	-	-
Base oil C (mass%)	-	-	77.5	-	-	-	-	29.0			-	-	-	-	-	-
Additive A (mass%)	10.0	10.0	10.0	20.0	5.0	3.0	10.0	10.0	10.0	10.0	-	-	-	-	-	-
Additive B (mass%)	-	-	-	-	-	-	-	-			-	10.0	-	-	-	-
Additive C (mass%)	-	-	-	-	-	-	-	-			-	-	10.0	-	-	-
Additive D (mass%)	-	-	-	-	-	-	-	-			-	-	-	10.0	-	-
Additive E (mass%)	-	-	-	-	-	-	-	-			-	-	-	-	10.0	-
Additive F (mass%)	-	-	-	-	-	-	-	-			-	-	-	-	-	10.0
Total (mass%)	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

(continued)

	Working Example										Comparative Example					
	1	2	3	4	5	6	7	8	9	10	1	2	3	4	5	6
Base Oil Kinematic Viscosity 100°C (mm <sup>2</sup> /s)	14.28	118.9	47.9	14.28	14.28	14.28	14.28	91.0	14.28	14.28	14.28	14.28	14.28	14.28	14.28	14.28
Base Oil Kinematic Viscosity 40°C (mm <sup>2</sup> /s)	144.9	15.4	8.2	144.9	144.9	144.9	144.9	12.0	144.9	144.9	144.9	144.9	144.9	144.9	144.9	144.9
Worked Penetration* (JIS K2220.7)	241	262	252	274	265	223	368	408	295	283	222	238	238	235	243	240
NLGI Consistency Grade	3	2.5	2.5	2	2	3	0	00	2	2	3	3	3	3	3	3
Dropping Point (JIS K2220.8) (°C)	265	254	255	257	267	265	223	224	270	182	258	243	247	266	272	274
High-speed Four Ball Wear Test: Wear scar diam. ** (mm)	0.39	0.42	0.43	0.45	0.44	0.46	0.43	0.46	0.50	0.52	0.59	0.63	0.66	0.65	0.67	0.57
* Worked penetration: 60W at 25°C (measured with a 1/2 penetration/consistency meter)																
** High-speed four ball wear test: 1,200rpm, 40kgf, ambient temperature (no temperature control), 30 min																

## Claims

1. A grease composition comprising a base oil, a thickener and poly(meth)acrylate which contains hydroxyl groups, wherein the thickener is a urea compound formed by the reaction between an isocyanate and primary amine; wherein the urea compound is a compound represented by general formula (I):



wherein  $R^1$  and  $R^3$  each represents a  $C_{8-12}$  aliphatic hydrocarbon group, and  $R^2$  represents a  $C_{6-15}$  divalent aromatic group; and wherein the thickener is present in an amount of from 1 to 20 mass%, taking the total grease composition as 100 mass%.

2. A grease composition according to claim 1, wherein the poly (meth) acrylate which contains hydroxyl groups is a copolymer having as constituent monomers an alkyl (meth) acrylate having a  $C_{1-20}$  alkyl group, and a hydroxyl-group-containing vinyl monomer.
3. A grease composition according to claim 2, wherein the alkyl (meth) acrylate having a  $C_{1-20}$  alkyl group is an alkyl (meth) acrylate having a  $C_{8-20}$  alkyl group.
4. A grease composition according to claim 2 or claim 3, wherein the hydroxyl-group-containing vinyl monomer is chosen from hydroxyalkyl ( $C_{2-6}$ ) (meth) acrylates, mono- or di-hydroxyalkyl ( $C_{1-4}$ ) -substituted (meth)acrylamides, vinyl alcohol formed by the hydrolysis of vinyl acetate units,  $C_{3-12}$  alkenols,  $C_{4-12}$  alkene diols, hydroxyalkyl ( $C_{1-6}$ ) alkenyl ( $C_{3-10}$ ) ethers, hydroxyl-group-containing aromatic monomers, polyhydric alcohols, and vinyl monomers containing a polyoxyalkylene chain and hydroxyl group(s).
5. A grease composition according to any one of claims 2 to 3, wherein the copolymer has further constituent monomers chosen from nitro-group-containing monomers, vinyl monomers containing primary, secondary or tertiary amino groups, amphoteric vinyl monomers and monomers containing a nitrile group.
6. A grease composition according to any preceding claim, wherein the hydroxyl value of the poly(meth)acrylate containing hydroxyl groups is from 20 to 50 measured based on JIS K3342 (1961).
7. A grease composition according to any preceding claim, comprising from 60 to 95 mass% of base oil, and from 2-20 mass% of the poly(meth)acrylate which contains hydroxyl groups, taking the total grease composition as 100 mass%.

## Patentansprüche

1. Fettzusammensetzung, umfassend ein Grundöl, ein Verdickungsmittel und Poly(meth)acrylat, das Hydroxylgruppen enthält, wobei das Verdickungsmittel eine Harnstoffverbindung ist, die durch die Reaktion zwischen einem Isocyanat und einem primären Amin ausgebildet wird; wobei die Harnstoffverbindung eine Verbindung ist, die durch eine allgemeine Formel (I) dargestellt wird:



wobei  $R^1$  und  $R^3$  jeweils eine aliphatische  $C_{8-12}$ -Kohlenwasserstoffgruppe darstellen und  $R^2$  eine zweiwertige aromatische  $C_{6-15}$ -Gruppe darstellt; und wobei das Verdickungsmittel in einer Menge von 1 bis 20 Masse-% vorhanden ist, wobei die gesamte Fettzusammensetzung als 100 Masse-% angenommen wird.

2. Fettzusammensetzung nach Anspruch 1, wobei das Poly(meth)acrylat, das Hydroxylgruppen enthält, ein Copolymer ist, das als konstituierende Monomere ein Alkyl (meth) acrylat mit einer  $C_{1-20}$ -Alkylgruppe und ein hydroxylgruppehaltiges Vinylmonomer aufweist.
3. Fettzusammensetzung nach Anspruch 2, wobei das Alkyl (meth) acrylat mit einer  $C_{1-20}$ -Alkylgruppe ein Alkyl (meth) acrylat mit einer  $C_{8-20}$ -Alkylgruppe ist.
4. Fettzusammensetzung nach Anspruch 2 oder 3, wobei das hydroxylgruppehaltige Vinylmonomer aus Folgenden

ausgewählt ist: Hydroxyalkyl(C<sub>2-6</sub>)-(meth)acrylaten, Mono- oder Dihydroxyalkyl (C<sub>1-4</sub>)substituierten (Meth)acrylamiden, Vinylalkohol, der durch die Hydrolyse von Vinylacetateinheiten ausgebildet wird, C<sub>3-12</sub>-Alkenolen, C<sub>4-12</sub>-Alkendiolen, Hydroxyalkyl (C<sub>1-6</sub>)alkenyl (C<sub>3-10</sub>) ethern, hydroxylgruppehaltigen aromatischen Monomeren, mehrwertigen Alkoholen und Vinylmonomeren, die eine Polyoxyalkylenkette und (eine) Hydroxylgruppe(n) enthalten.

- 5 5. Fettzusammensetzung nach einem der Ansprüche 2 bis 3, wobei das Copolymer weitere konstituierende Monomere aufweist, die aus Folgenden ausgewählt sind: nitrogruppehaltigen Monomeren, Vinylmonomeren, die primäre, sekundäre oder tertiäre Aminogruppen enthalten, amphoteren Vinylmonomeren und Monomeren, die eine Nitrilgruppe enthalten.
- 10 6. Fettzusammensetzung nach einem vorhergehenden Anspruch, wobei die Hydroxylzahl des Poly(meth)acrylats, das Hydroxylgruppen enthält, basierend auf JIS K3342 (1961) gemessen, von 20 bis 50 beträgt.
- 15 7. Fettzusammensetzung nach einem vorhergehenden Anspruch, umfassend von 60 bis 95 Massen-% Grundöl und von 2-20 Massen-% des Poly(meth)acrylats, das Hydroxylgruppen enthält, wobei die gesamte Fettzusammensetzung als 100 Massen-% angenommen wird.

## Revendications

- 20 1. Une composition de graisse comprenant une huile de base, un épaississant et un poly(méth)acrylate qui contient des groupes hydroxyles, dans laquelle l'épaississant est un composé urée formé par la réaction entre un isocyanate et une amine primaire ;  
dans laquelle le composé urée est un composé représenté par la formule générale (I) :



dans laquelle R<sup>1</sup> et R<sup>3</sup> représentent chacun un groupe hydrocarboné aliphatique en C<sub>8</sub> à C<sub>12</sub>, et R<sup>2</sup> représente un groupe aromatique divalent en C<sub>6</sub> à C<sub>15</sub> ; et

30 dans laquelle l'épaississant est présent en une quantité de 1 à 20 % en masse, amenant la composition totale de graisse à 100 % en masse.

- 35 2. Composition de graisse selon la revendication 1, dans laquelle le poly(méth)acrylate qui contient des groupes hydroxyles est un copolymère ayant comme monomères constitutifs un (méth)acrylate d'alkyle ayant un groupe alkyle en C<sub>1</sub> à C<sub>20</sub>, et un monomère vinylique contenant un groupe hydroxyle.
- 40 3. Composition de graisse selon la revendication 2, dans laquelle le (méth)acrylate d'alkyle ayant un groupe alkyle en C<sub>1</sub> à C<sub>20</sub> est un (méth)acrylate d'alkyle ayant un groupe alkyle en C<sub>8</sub> à C<sub>20</sub>.
- 45 4. Composition de graisse selon la revendication 2 ou la revendication 3, dans laquelle le monomère vinylique contenant un groupe hydroxyle est choisi parmi les (méth)acrylates d'hydroxyalkyle (C<sub>2</sub> à C<sub>6</sub>), les (méth)acrylamides mono- ou di-hydroxyalkyle (C<sub>1</sub> à C<sub>4</sub>)-substitués, l'alcool vinylique formé par l'hydrolyse de motifs d'acétate de vinyle, les alcénols C<sub>3</sub> à C<sub>12</sub>, les alcènes diols C<sub>4</sub> à C<sub>12</sub>, l'hydroxyalkyle (C<sub>1</sub> à C<sub>6</sub>) les éthers d'alcényle (C<sub>3</sub> à C<sub>10</sub>), les monomères aromatiques contenant un groupe hydroxyle, les polyols et les monomères vinyliques contenant une chaîne polyoxyalkylène et un ou des groupes hydroxyle(s).
- 50 5. Composition de graisse selon l'une quelconque des revendications 2 à 3, dans laquelle le copolymère a d'autres monomères constitutifs choisis parmi les monomères contenant des groupes nitro, les monomères vinyliques contenant des groupes amino primaires, secondaires ou tertiaires, les monomères vinyliques amphotères et les monomères contenant un groupe nitrile.
- 55 6. Composition de graisse selon l'une quelconque des revendications précédentes, dans laquelle l'indice d'hydroxyle du poly(méth)acrylate contenant des groupes hydroxyles est de 20 à 50, mesuré selon la norme JIS K3342 (1961).
7. Composition de graisse selon l'une quelconque des revendications précédentes, comprenant de 60 à 95 % en masse d'huile de base, et de 2 à 20 % en masse du poly(méth)acrylate qui contient des groupes hydroxyles, amenant la composition de graisse totale à 100 % en masse.

**REFERENCES CITED IN THE DESCRIPTION**

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