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54 **A lubricating oil composition.**

57 The lubricating oil composition of the present invention comprising an ester mixture as the main ingredient consisting of (A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms, the composition having excellent performances such as biodegradability, high-temperature engine cleanliness, lubricity and piston seizure preventiveness which are conducive to environmental protection.

EP 0 535 990 A1

The present invention relates to a lubricating oil composition and more particularly to a lubricating oil composition which is excellent in biodegradability, high-temperature cleanliness and anti-seizure performance and is therefore conducive to environmental protection.

Two-cycle engines raise problems as to possible environmental pollution since they discharge an unburned engine oil as an accompaniment of their exhaust gases due to their lubrication mechanism. For example, outboard engines used in rivers, lakes or oceans discharge exhaust gases containing unburned oil into the water thereby to raise problems as to water pollution due to the unburned oil. Chain saws and other forestry machinery may also cause the pollution of forests and rivers due to the unburned oil.

In Europe, there have been lively activities in attempts to establish laws, regulations and standards concerning the biodegradability of the two-cycle engine oils from the standpoint of environmental protection. Because of this, a two-cycle engine oil having biodegradability has heretofore been tried to be developed and it has already been sold from several oil makers. These oils, in all cases, comprise, as a specific base oil an ester compound (based on a mixture of saturated and unsaturated fatty acid esters each having 16 - 18 carbon atoms) and a mineral oil-based solvent. They contain an amino amide type ashless dispersant as an additive.

These commercially available oils already passed the TC-WII which is a performance standard of oils for outboard engines. However, when these oils are used in recently manufactured high-performance water-cooled outboard engines and air-cooled engines, they are considered to raise problems as to ring sticking and piston seizure due to their insufficient thermal stability as a lubricating oil.

Because of this, a biodegradable lubricating oil having better thermal stability and more excellent lubricity has been sought to be developed.

An object of the present invention is to provide a lubricating oil having excellent performances in biodegradability, high-temperature cleanliness, lubricity and anti-seizure performance.

Another object is to provide a method for lubrication comprising the use of such a lubricating oil as above.

The present inventors made intensive studies to aim mainly at solving the above problems and, as the result of their studies, they found out that a lubricating oil comprising predetermined esters as main components satisfies the above-mentioned requirements, thus achieving the present invention.

More particularly, in a first aspect of the present invention, a lubricating oil composition comprises as the main component an ester mixture consisting of (A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms.

In a second aspect of the present invention, a lubricating oil composition comprises (I) 100 parts by weight of an ester mixture consisting essentially of (A) 60 - 95 % by weight of an ester of a hindered alcohol and a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with both a straight-chain saturated fatty acid having 8 - 12 carbon atoms and a dibasic acid having 2 - 50 carbon atoms, and (II) not more than 30 parts by weight of a hydrocarbon-based solvent and/or a lubricating base oil.

The present invention will now be explained hereunder in more detail.

The ester mixture used in the present invention as the main ingredient is a mixture of (A) an ester of a hindered alcohol and a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and (B) a complex ester of a hindered alcohol with both a straight-chain saturated fatty acid having 8 - 12 carbon atoms and a dibasic acid having 2 - 50 carbon atoms.

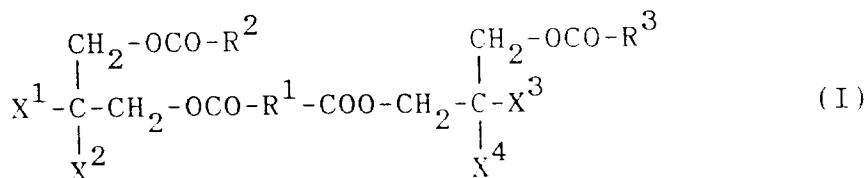
In the preparation of this component (A), the hindered alcohol used means one in which the β carbon atom of the hydroxyl group is a quarternary carbon atom. More specifically, the hindered alcohol preferably used include dihydric to tetrahydric alcohols having 5 - 10 carbon atoms, and a dimer and trimer thereof, and they are exemplified by neopentyl glycol, 2-methyl-2-propyl-1, 3- propanediol, trimethylol ethane, trimethylol propane (TMP), trimethylol butane, pentaerythritol (PET), di- (trimethylol propane), tri-(trimethylol propane), di- (pentaerythritol) and tri-(pentaerythritol).

The straight-chain saturated fatty acid having 8 - 12 carbon atoms used in the present invention include caprylic acid, pelargonic acid, capric acid and lauric acid. Furthermore, such fatty acids may be used in the form of a derivative such as an acid anhydride, acid halide and metal salt of the acids. In the preparation of the component (A), at least two different acids among these fatty acids may be esterified with the same hindered alcohol, or the resulting ester may have a hydroxyl group therein which remains without being esterified.

In the preparation of the component (B), the same hindered alcohol and the saturated fatty acid having 8 - 12 carbon atoms as those used in the preparation of the above component (A) may be employed. The

dibasic acids having 2 - 50 carbon atoms used in said preparation include oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid and sebacic acid, as well as other acids having more carbon atoms than the above-mentioned acids. Among these acids, those having 6 - 40 carbon atoms are desirably used.

The component (B) may be exemplified by a complex ester represented by the following general formula (I).



wherein X¹ to X⁴ may be identical with or different from each other and each represent an alkyl group having 1 - 4 carbon atoms or a group having the following general formula of -CH₂-OCO-R⁴ (R⁴ being an alkyl group having 7 - 11 carbon atoms). Also, in the formula (I), R¹ represents an alkylene or alkenylene group having 1 - 48 carbon atoms with the proviso that when the number of carbon atoms of a dibasic acid is 2, R¹ does not exist in the formula I. R² to R⁴ may be identical with or different from each other and each represent a straight-chain alkyl group having 7 - 11 carbon atoms. In this case, R¹ corresponds to the alkylene group of a dibasic acid having 3 - 50 carbon atoms, R² to R⁴ each correspond to the alkyl group of a straight-chain saturated fatty acid having 8 - 12 carbon atoms.

In this invention, the content of the above component (A) is 60 to 95%, preferably 70 to 90%, by weight based on the total weight of the specific base oil. The content of the above component (B) is 5 to 40%, preferably 10 to 30%, by weight based on the total weight of the specific base oil. When the contents of the above components (A) and (B) do not fall respectively within the above ranges, the resulting composition will not be appropriate in viscosity. In addition, when the content of the above component (B) exceeds 40% by weight, the resulting composition will undesirably be insufficient in biodegradability. Furthermore, the composition of the present invention should have a kinematic viscosity of preferably 6 to 15 mm²/s at a temperature of 100 °C.

The lubricating oil composition of the present invention may contain the above ester mixture alone, and, however, it may further contain a known hydrocarbon-based solvent and/or a known lubricating base oil as required. This hydrocarbon-based solvent may be one which is usually used for two-cycle engine oils, and it is exemplified by a petroleum-based hydrocarbon solvent and/or a synthesized hydrocarbon solvent each having a boiling point of 150 to 300 °C at atmospheric pressure. Specifically, these solvents are illustrated by Stoddard solvent, mineral spirits, kerosene fractions, n-paraffins, i-paraffins and propylene oligomers.

The lubricating base oil may be one which is usually used as a base oil for lubricating oils and it is exemplified by paraffinic and naphthenic mineral oils prepared by refining lubricating oil fractions which are obtained by distilling crude oils under atmospheric or a reduced pressure; poly- α -olefin (1-octene oligomer, 1-decene oligomer, etc.); polybutene, alkylbenzenes; alkylnaphthalenes; polyglycol; diester (ditridecylglutarate, di,2-ethylhexyl adipate); diisodecyl adipate; di, tridecyl adipate; di,2-ethyl hexyl sebacate; esters of polyol with a straight-chain chain fatty acid having up to 7 or at least 13 carbon atoms or with a branched-chain fatty acid (trimethylolpropane stearate, trimethylolpropane oleate, pentaerithritol 2-ethylhexanoate, etc.); polyphenylether; fluorine-based oils; and silicone-based oils.

In a case where the solvent and/or a lubricating base oil other than the above-mentioned ester mixture (the specific base oil) are/is to be added to the ester mixture, it is preferable to add the solvent and/or the lubricating base oil in a total amount of up to 30 parts, preferably up to 20 parts, by weight per 100 parts by weight of the specific base oil consisting of the ester mixture so as not to deteriorate the excellent thermal stability, lubricity and also biodegradability which are the features of this invention.

Moreover, the composition of this invention may be incorporated with various kinds of known additives for the purpose of further improving the performance of the composition, as required. The additives include basic calcium sulfonate, basic calcium phenate, basic calcium salicylate, alkenyl succinic acid imide, benzyl amine, a detergent such as a polyalkenyl amine, a pour point depressant such as polymethacrylate, a rust preventing agent and anti-foaming agent.

These additives may be added alone or jointly. They may be added in any optional amount and, usually, they may each be added in an amount of up to 30, preferably 0.5 - 15, parts by weight per 100 parts by weight of the specific base oil.

Although the lubricating oil compositions of this invention are suited especially for two-cycle engines such as outboards and chain saws since they are excellent in biodegradability, they are preferably used in engines for two-wheeled vehicles such as mopet (motorlike) and motorcycles, and in portable power unit engines for lawnmowers and power generators. Furthermore, they can be used as a four-cycle engine oil, a hydraulic oil, a gear oil and a metal processing oil.

The present invention will be better understood by way of Examples and Comparative examples, and, however, it is not limited to these Examples. 1. biodegradability and thermal stability

The biodegradability and thermal stability which are fundamental performances of this invention were evaluated.

(1) biodegradability

The biodegradability was measured by the coulometer method using a closed-system oxygen consumption measurement apparatus, and this method is usually called the MITI method which is one of the test methods prescribed by the Chemical Substances Control Law in Japan. This method is the one in which incubation is effected at a temperature of 25 ° C for a period of time of 14 days.

A decomposition degree is represented by the following formula.

$$\text{Decomposition degree} = [(BOD-B)/TOD] \times 100$$

BOD: Biological Oxygen Demand of a test substance (Value found, mg)

B: Oxygen consumption of a culture medium into which activated sludge has been inoculated (Value found, mg)

TOD: Theoretical Oxygen Demand required for complete oxidation of the test substance (Value calculated, mg). The value for TOD was determined by calculating the molecular formula obtained from the elemental analysis of the test substance (oil).

A test substance which showed a decomposition degree of 35 to 40 % in this test is considered to have undergone almost complete biodegradation, and, therefore, the biodegradability standard for the compositions of this invention has been determined in conformity with said decomposition degree.

(2) Thermal stability

Thermal stability was evaluated by a hot tube test (HTT) (This test is described in SAE Paper 881619, 1988). This test was carried out by installing a glass tube into an electric furnace and then pushing a test oil upward with air through the glass tube. The oil was degraded by being subjected to heat and oxidation when it passed through the glass tube. In this test, a merit rating is represented by the shade of lacquer-like color of a deposit to the inner wall of the glass tube. The merit rating is between 10 points when no deposit is found, and 0 point when a deposit looks black in color. Because there is comparatively good correlation between piston cleanliness found by engine tests and HTT merit rating, the HTT is utilized as a screening test before subjecting to an engine test. The compositions of this invention were produced so that they would exhibit a HTT merit rating of at least 5 points at 280 ° C, 16 hrs as their standard.

The test results thus obtained are shown in Table 1.

T a b l e 1

Example Comp. Example	Component (A) wt. %		Component (B) wt. %		Other oils, parts by weight based on 100 parts by weight of (A) + (B)	Additives, parts by weight based on 100 parts by weight of (A) + (B)	Test result of biodegradability (MTI method) decomposition degree %	Test result of HTT at 280°C. for 16 hrs. the best = 10, the worst = 0
	A-1	A-2	B-1	B-2				
Ex. 1	95	-	5	-	0	ashless dispersant A 20	39	7
Ex. 2	60	-	-	40	diluent 5	ashless dispersant B 15	40	8
Ex. 3	-	80	20	-	diluent 10	ashless dispersant B 15	45	8
Ex. 4	80	-	20	-	diluent 5	ashless dispersant B 15	51	7
Ex. 5	-	70	-	30	mineral oil 5 (SAE30)	ashless dispersant C 5 metallic detergent 1	39	7
Ex. 6	-	95	-	5	diluent 5	ashless decomposer B 25	38	5
Comp. Ex. 1	100 wt. % of TMP ester consisting mainly of fatty acid having 18 carbon atoms						45	3
Comp. Ex. 2	100 wt. % of PET ester consisting of fatty acid having 8 carbon atoms and iso-structure						3	10

Notes:
 ashless dispersant A: bis type succinimide
 ashless dispersant B: benzyl amine + aminoamide
 ashless dispersant C: bis type succinimide
 metallic detergent : basic calcium sulfonate

T a b l e 1 (Continued)

Comp. Ex.	Component (A) wt. %		Component (B) wt. %		Other oils, parts by weight based on 100 parts by weight of (A) + (B)	Additives, parts by weight based on 100 parts by weight of (A) + (B)	Test result of biodegradability (MITI method) decomposition degree %	Test result of HTT at 280°C. for 16 hrs. the best = 10, the worst = 0
	A-1	A-2	B-1	B-2				
Comp. Ex. 3	(A-1) 100 wt. %						45	10
Comp. Ex. 4	(A-2) 100 wt. %						40	10
Comp. Ex. 5	(B-1) 100 wt. %						24	10
Comp. Ex. 6	(A-1) 50 wt. %, (B-1) 50 wt. %						30	10
Comp. Ex. 7	commercially available biodegradable two-cycle engine oil-1						43	0
Comp. Ex. 8	commercially available biodegradable two-cycle engine oil-2						40	0
Comp. Ex. 9	commercially available low-smoke exhaust gas type two-cycle engine oil-1						8	10
Comp. Ex. 10	commercially available mineral base oil for two-cycle engine oil						14	3
Comp. Ex. 11	commercially available low-smoke exhaust gas type two-cycle engine oil-2						10	3

Notes:

- (A-1) : TMP ester of saturated fatty acid having 8-12 carbon atoms (mean molecular weight : 600)
 (A-2) : PET ester of saturated fatty acid having 8-12 carbon atoms (mean molecular weight : 800)
 (B-1) : TMP complex ester consisting of saturated fatty acid having 8-12 carbon atoms and dibasic acid having 36 carbon atoms (mean molecular weight : 1300)
 (B-2) : PET complex ester consisting of saturated fatty acid having 8-12 carbon atoms and dibasic acid having 36 carbon atoms (mean molecular weight : 1300)
 diluent : kerosine

Examples 1 to 6 which show compositions of this invention, exhibited excellent performances in biodegradability and thermal stability as compared with Comparative Examples which show comparative compositions.

(1) Comparative Example 1 is a TMP ester which consists mainly of a fatty acid (oleic acid) having 18 carbon atoms. This ester exhibited good biodegradability, and, however, it was poor in thermal stability

as compared the composition of the present invention.

(2) Comparative Example 2 is a PET ester which consists of a branched-chain fatty acid having 8 carbon atoms. This PET ester exhibited low biodegradability although it exhibited good thermal stability.

(3) Comparative Examples 3 and 4 used therein oils alone which were the same as the components (A-1) and (A-2) of this invention, respectively. Each of these oils exhibited good biodegradability and thermal stability. However, in a case where each of said oils is used alone as a lubricating oil, it exhibits a kinematic viscosity of less than 6 mm²/s at 100 °C. This viscosity is too low as a base oil for a two-cycle engine oil, and therefore, these oils are likely to exhibit insufficient lubricity and insufficient anti-seizure performance. In addition, Comparative Example 5 used therein an oil alone which was the same as the component (B) of this invention. Unlike the oils in Comparative Examples 3 and 4, the oil alone in Comparative Example 5 exhibited a kinematic viscosity of higher than 30 mm²/s at 100 °C, and raised problems as to its detergency and fluidity.

(4) Comparative Example 6 used therein an oil prepared by mixing the components (A) and (B) of this invention. The oil so prepared exhibited inferior biodegradability since the mixing ratio was outside that specified for the composition of this invention.

(5) Comparative Examples 7 and 8 show commercially-available biodegradable two-cycle engine oils respectively. These oils use therein the same ester as used in Comparative Example 1 as a main component of the base oil. In contrast with these Comparative Examples, it is apparent that the compositions of this invention shown in Examples 1 to 6 were remarkably improved in thermal stability. In addition, Comparative Example 9 shows a polybutene base oil for commercially-available low-smoke exhaust gas type two-cycle engine oil. This polybutene base oil exhibits good thermal stability in the HTT, but it has low biodegradability. Comparative Example 10 shows a mineral base oil for two-cycle engine oil, and Comparative Example 11 shows an oil for commercially-available low-smoke exhaust gas type two-cycle engine oil, in which the base oil has partly been replaced with a polybutene base oil. Such a mineral base oil is poor in both thermal stability and biodegradability.

(6) As shown in the Examples, the composition of this invention comprises the components (A) and (B) in the specified mixing ratio thereby enabling the composition to have a suitable range of viscosity which is required in two-cycle engine oils, and also to exhibit excellent lubricity as well as excellent biodegradability and thermal stability which are important properties of the composition of this invention.

2. Engine test results

(1) High-temperature cleanliness test on motorcycle engines

Using an air-cooled two-cycle, 1-cylinder engine having a displacement of 123 c.c. for motorcycles, a high-temperature cleanliness test was carried out under conditions of an engine speed of 7000rpm, full engine load, a plug gasket temperature of 260 °C, a fuel oil mixing ratio of 20:1 and testing time of 3hrs.

As shown in Table 2, the composition of this invention in Example 3 was excellent in anti-seizure performance between the piston and the cylinder as compared with Comparative Examples 7 and 8 which were commercially-available biodegradable two-cycle engine oils. This composition of Example 3 was also extremely excellent in piston cleanliness without ring sticking after the test.

Table 2

Test oils		Ex. 3	Comp. Ex. 7	Comp. Ex. 8
ring sticking,	top ring	10	0	6
	second ring	10	0	6
ring land,	top	4.1	1.5	3.8
	second	5.6	0.3	4.1
piston skirt		9.4	5.3	9.2
undercrown		2.9	0.6	2.0
Total: 60 points (full marks)		42.0	7.7	33.1
Note: Since the piston seizure took place in 2.5 hours in Comparative Example 7, Comparative Example 7 shows the piston cleanliness at that time.				

(2) Cleanliness test on engine for electric generator

Using a 1-cylinder engine employed for a generator and having a displacement of 63 cc, an engine cleanliness test was made under conditions of full engine load (800 W) and testing time of 5hrs. A plug gasket temperature was set at 200 °C by covering about 50% of the air intake of a forced cooling fan. The test was carried out at a fuel:oil mixing ratio of 50 : 1 (mixing lubrication).

As shown in Table 3, the composition of Example 4 according to this invention was excellent in piston cleanliness without causing ring sticking as compared with Comparative Example 7 which was a commercially available biodegradable two-cycle engine oil and with Comparative Example 8 which has a mineral base oil. The composition of Example 4 was also extremely low in cylinder head deposits.

Table 3

Test oils		Ex. 4	Comp. Ex. 7	Comp. Ex. 10
ring sticking,	top ring	10	8	10
	second ring	10	10	10
ring land,	top	6.7	1.0	1.6
	second	8.2	5.3	4.4
piston skirt		10	9.3	9.2
undercrown		10	2.2	1.6
cylinder head		10	8.9	5.7
Total: 70 points (full marks)		64.9	44.7	42.5

(3) Engine cleanliness test on chain saw engine

An engine cleanliness test was carried out on a 1-cylinder engine used for a chain saw engine and having a displacement of 45 cc, under conditions of an engine speed of 9000rpm, full engine load, a plug gasket temperature of 280 °C, a fuel oil mixing ratio of 50:1 and testing time of 30 hrs (mixing lubrication).

As shown in Table 4, the composition of Example 4 according to this invention was extremely excellent in ring sticking resistance and piston cleanliness as compared with Comparative Example 11 which was an oil for a commercially-available low-smoke type two-cycle engine oil.

Table 4

Test oils		Ex. 4	Comp. Ex. 11
ring sticking,	top ring	7.0	5.0
	second ring	10	10
ring land,	top	8.4	3.9
	second	9.5	5.2
piston skart		10	9.0
undercrown		2.4	0.9
Total: 60 points (full marks)		47.3	34.0

As described above, several points of excellent performances of the composition according to this invention were illustrated in the tests, and, further, the same results were obtained even in other engine tests. In addition, the composition of this invention has already passed the TC-WII which is a standard of NMMA (National Marine Manufacturers Association) for two-cycle engine oils for use in outboard engines and has also passed the CEC L-33-T-82 which is a test method of evaluating biodegradability of two-cycle engine oils for outboard engines (the method of biodegradability test is different from the MITI method of Japan) and which indicates that a standard of the biodegradability is at least 67 % (for example, Example 4 exhibited a decomposition degree of 87 %, and, on the other hand, Comparative Example 7 showed 67 % in this test method).

As will be understood from the above, the composition according to this invention is a lubricating oil which can solve the problems as to the piston ring sticking and piston seizure which have become problems in the market.

Claims

1. A lubricating oil composition comprising an ester mixture as the main ingredient consisting of
 - (A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and
 - (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms.
2. A lubricating oil composition comprising
 - (I) 100 parts by weight of an ester mixture consisting of
 - (A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and
 - (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms,
 - (II) not more than 30 parts by weight of a hydrocarbon based solvent and/or a lubricating base oil.
3. A lubricating oil composition according to claim 1, which is used as a two-cycle engine oil.
4. A lubricating oil composition according to claim 2, which is used as a two-cycle engine oil.
5. A method for lubrication comprising the use of a lubricating oil composition comprising an ester mixture as the main ingredient consisting of
 - (A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms, and
 - (B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms.
6. A method for lubrication comprising the use of a lubricating oil composition comprising
 - (I) 100 parts by weight of an ester mixture consisting of

(A) 60 - 95 % by weight of an ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and

(B) 5 - 40 % by weight of a complex ester of a hindered alcohol with a straight-chain saturated fatty acid having 8 - 12 carbon atoms and also with a dibasic acid having 2 - 50 carbon atoms, and

(II) not more than 30 parts by weight of a hydrocarbon based solvent and/or a lubricating base oil.

7. A method for lubrication according to claim 5 or 6, which is applied to a two-cycle engine.



European Patent
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EUROPEAN SEARCH REPORT

Application Number

EP 92 30 9033

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	FR-A-2 144 559 (I.F.P.) * page 3, line 33 - page 4, line 9; example 3 * ---	1-7	C10M105/32 C10M169/04 C10M111/02
X	US-A-3 505 230 (THOMPSON) * column 4, line 33 - line 47 * * column 7, line 15 * * column 8, line 21 - line 23 * ---	1,5	
A	DE-A-2 551 173 (HENKEL) * claims 1,12 * ---	1-7	
A	FR-A-2 311 088 (I.F.P.) * claims 1-6 * ---	1-7	
A	EP-A-0 435 253 (NIPPON OIL) * claim 1 * ---	1-7	
A	EP-A-0 410 170 (SCHUR ET AL.) * column 2, line 31 * ---	1-7	
A	US-A-3 218 256 (EDWARDS ET AL.) * column 3 - column 5 * -----	1-7	TECHNICAL FIELDS SEARCHED (Int. Cl.5) C10M
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 16 DECEMBER 1992	Examiner DE LA MORINERIE
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document			