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(54) **Lubricating oil additive concentrate composition.**

(57) A concentrate composition for use as a multifunctional additive in finished lubricating oils comprising as a first component an oil of lubricating viscosity, as a second component a derivative of a polyisobutylene (PIB) substituted succinic acylating agent wherein the PIB substituent has a number average molecular weight (Mn) in the range from about 700 to about 1475, as a third component a derivative of a PIB substituted succinic acylating agent wherein the PIB has an Mn in the range from about 1500 to about 5000 and as a fourth component a PIB having an Mn in the range from about 40,000 to about 400,000, the derivatives being formed by reacting the acylating agents either individually or in combination with either (a) an amine having within its structure at least one H-N group, (b) an alcohol, (c) a reactive metal or reactive metal compound, or (d) a mixture of at least two of (a) to (c), the components of (d) being reacted with the acylating agent(s) either simultaneously or sequentially in any order, the PIB substituent of the second component forming from 30 to 94.8 % by weight of the total PIB present in the composition, the PIB substituent of the third component forming from 5 to 69.8% by weight of the total PIB present in the composition and the PIB forming the fourth component forming from 0.2 to 5% by weight of the total PIB present in the composition.

EP 0 342 871 A1

LUBRICATING OIL ADDITIVE CONCENTRATE COMPOSITION

The present invention relates generally to lubricating oil additive concentrate compositions and their use in finished lubricating oil compositions. In particular the present invention relates to lubricating oil additive concentrate compositions having both dispersancy/detergency properties and viscosity improving properties and their use in finished lubricating oil compositions.

5 It is known that polyisobutylenes of medium to high molecular weight, for example weight average molecular weights above about 10,000, typically greater than 40,000, act as viscosity index improvers in lubricating oil compositions. Such viscosity index improvers, for example polyisobutylenes having average molecular weights of 60,000 to 80,000 or more, exhibit virtually no dispersant or detergent properties. That is, they are monofunctional additives imparting to the lubricant only the desired viscosity index improving
10 properties. To obtain dispersant or detergent properties, such hydrocarbon viscosity index improvers are used in combination with one or more dispersant or detergent additives.

It is also known that polyisobutylenes can be converted into detergent/dispersant additives for lubricating oil compositions. Thus, for example, the patent literature discloses the preparation of high molecular weight carboxylic acid acylating agents by reacting a polyisobutylene usually containing at least about 50
15 aliphatic carbon atoms with an unsaturated carboxylic acid or a derivative thereof, for example acrylic acid, fumaric acid and maleic anhydride, see for example US-A-3,024,237; US-A-3,087,936; US-A-3,172,892; US-A-3,215,707; US-A-3,219,666; US-A-3,231,587; US-A-3,245,910; US-A-3,272,746; GB-A-1,085,903; GB-A-1,162,436, GB-A-1,440,219 and many more, the foregoing only being representative of the many patent publications in this area. These same patents also establish that various derivatives, for example the
20 succinimide derivatives, of these high molecular weight carboxylic acid acylating agents are known to be useful as additives in fuel and lubricant compositions, especially as dispersant/detergent additives, which function to promote engine cleanliness, neutralise acidic by-products of combustion, and the like. Some of the compositions disclosed in the above patents have been used in substantial amounts as commercial lubricant additives. The aforesaid derivatives of high molecular weight acylating agents exhibit substantially
25 no viscosity index improver properties. That is they too are essentially monofunctional additives imparting to the lubricant only the desired dispersancy and detergency properties. To obtain viscosity index improver properties such dispersant/detergent additives are generally used in combination with a viscosity index improver additive.

More recently, there has been a move towards multifunctional additives, that is a single additive which
30 exhibits both viscosity index improver and dispersant/detergent properties. In general, two approaches have been used to prepare multifunctional lubricant additives which exhibit both (a) fluidity modifying properties, especially viscosity index improver properties, and (b) dispersant and/or detergent properties. One approach involves "suspending" from or "incorporating" into the hydrocarbon backbone of a high molecular weight polymer certain polar groups (usually carboxylic acid derivatives such as amides and esters). The
35 high molecular weight material so-produced continues to exhibit viscosity index improver properties attributable to its high molecular weight hydrocarbon backbone and dispersant/detergent properties attributable to the polar groups. This approach is illustrated by US-A-3,702,300 and US-A-3,933,761. The second general approach for preparing multifunctional lubricant additives involves modifying a dispersant/detergent additive so as to incorporate into the dispersant/detergent additive fluidity modifying properties, especially
40 viscosity index improver properties. This approach is illustrated by US-A-3,219,666. This patent is mainly concerned with acylated nitrogen derivatives of high molecular weight succinic acid acylating agents, which derivatives function as dispersant additives in lubricant compositions. The acylating agents from which the dispersants are prepared are substituted succinic acylating agents preferably having a substituent derived from a polyisobutylene having a molecular weight of about 750-5000. However, the patent teaches that, if
45 viscosity index improving properties are desired in addition to the dispersant properties, the substituent should be derived from higher molecular weight polyisobutylene polymers, having molecular weights from about 10,000 to about 100,000 or higher. A third approach for preparing lubricant additives having both detergency and viscosity index-improving properties is described in US-A-3,630,902. This approach involves reacting a high molecular weight succinimide with a polymerisable acid to form a polymerisable
50 acyl derivative of the succinimide. The polymerisable derivative is then polymerised to produce the desired multifunctional lubricant additive.

A further approach to the production of a multifunctional lubricating oil additive is described in GB-A-1,565,627. This patent provides a lubricating composition comprising a major amount of oil of lubricating viscosity and a minor amount of one or more carboxylic derivatives produced by reacting at least one substituted succinic acylating agent with a reactant selected from (a) an amine having within its structure at

least one H-N< group, (b) an alcohol, (c) a reactive metal or reactive metal compound, and (d) a combination of two or more of any of (a) to (c), the components of (d) being reacted with said one or more substituted succinic acylating agents simultaneously or sequentially in any order, wherein said substituted succinic acylating agent(s) consist of substituent groups and succinic groups wherein the substituent groups
 5 are derived from polyalkene, preferably polyisobutylene, said polyalkene having an Mn value of 1300 to 5000 and a Mw/Mn value of 1.5 to 4, said acylating agent(s) having within their structure an average of at least 1.3 succinic groups for each equivalent weight of substituent groups.

The only method described for producing the substituted acylating agents of GB-A-1,565,627 is via a chlorination route, which can result in chlorine residues in the final product. The presence of chlorine is
 10 undesirable because it can have deleterious effects in engines in which the finished lubricating oil is employed.

Lubricating oil additive packages are normally manufactured by an additive producer and sold on for blending into finished lubricating oils. The additive package generally consists of a mixture of dispersants, detergents, extreme pressure/anti-wear agents, rust inhibitors and other additives for special applications.
 15 This package can be used at levels of between 1% and 25% in oil in the automotive field, and up to 40% in the marine field. In addition in the automotive field a viscosity index improver is added if multigrade performance is required. The addition of viscosity index improvers (high molecular weight) to the normal package results in precipitation of the viscosity index improver (VII) rubber and/or the other components in the package. This is due to lack of solvent base oil in the additive carrier oil. This precipitation can be
 20 instantaneous or it can occur over a period of 1 day to 180 days or longer. However long the period over which precipitation occurs, it is clearly undesirable. The VII can be a hydrocarbon VII of the ethylene/propylene (EP) copolymer, ethylene/propylene/diene (EPDM), polyisobutene (PIB), polystyrene, hydrogenated polyisoprene, hydrogenated polystyrene/butadiene types etc, or it can be a dispersant polymer such as a polymethacrylate, a dispersant EP, a dispersant EPDM or a dispersant PIB.

In the present invention we have achieved by advantageous blending of PIBs a dispersant which not only has all the dispersant properties of a normal dispersant but also has a small VII contribution to the finished oil and most importantly it has complete compatibility with all of the other additives in a typical additive package at conventionally used levels. The blending of the PIBs can take place either before maleanisation and imidation and/or after maleanisation and imidation or at any time during the respective
 30 processes.

The use of mixtures of derivatives of polyisobutene-substituted succinic acylating agents in which the polyisobutene substituents have different molecular weights as additives is known in relation to fuels. Thus, GB-A-1,398,067 discloses a detergent additive for a hydrocarbon distillate fuel comprising a mixture of a high molecular weight hydrocarbyl amine and a low molecular weight hydrocarbyl amine, the high
 35 molecular weight hydrocarbyl amine containing at least one hydrocarbyl group having an average molecular weight in the range of from 1900 to 5000 and the low molecular weight hydrocarbyl amine containing at least one hydrocarbyl group having an average molecular weight in the range from 300 to 600, the weight ratio of low molecular weight hydrocarbyl amine to high molecular weight hydrocarbyl amine being in the range 0.5:5:1. Although primarily directed to hydrocarbon distillate fuel usage, it is mentioned in the
 40 specification that the mixture of high and low molecular weight hydrocarbyl amines may also be used as dispersants and detergents in lubricating oils. Such a mixture, first of all in relation to the low molecular weight hydrocarbyl amine has a hydrocarbyl group of a lower molecular weight than the lowest molecular weight useful in the composition of the present invention, and secondly lacks an essential property of the compositions of the present invention in that there is no component present of a molecular weight
 45 sufficiently high to impart adequate viscosity index improver properties to the composition.

According to the present invention there is provided a concentrate composition for use as a multifunctional additive in finished lubricating oils which composition as a first component comprises an oil of lubricating viscosity, as a second component a derivative of a polyisobutylene-substituted succinic acylating agent wherein the polyisobutylene substituent has a number average molecular weight in the range from
 50 about 700 to about 1475, as a third component a derivative of a polyisobutylene-substituted succinic acylating agent wherein the polyisobutylene has a number average molecular weight in the range from about 1500 to about 5000 and as a fourth component a polyisobutylene having a number average molecular weight in the range from about 40,000 to about 400,000, the derivatives being formed by reacting the acylating agents either individually or in combination with either (a) an amine having within its structure at
 55 least one H-N group, (b) an alcohol, (c) a reactive metal or reactive metal compound, or (d) a mixture of at least two of (a) to (c), the components of (d) being reacted with the acylating agent(s) either simultaneously or sequentially in any order, the polyisobutylene substituent of the second component forming from 30 to 94.8% by weight of the total polyisobutylene present in the composition, the polyisobutylene substituent of

the third component forming from 5 to 69.8% by weight of the total polyisobutylene present in the composition and the polyisobutylene forming the fourth component forming from 0.2 to 5% by weight of the total polyisobutylene present in the composition.

The concentrate composition may simply be formed by mixing the individual components in the lubricating oil. However, it is preferred to produce a mixture of the second and third components by the steps of:-

(I) reacting a mixture of a polyisobutylene having a number average molecular weight in the range from about 700 to about 1475 and a polyisobutylene having a number average molecular weight in the range from about 1500 to about 5000 with an unsaturated carboxylic acid or anhydride thereof to produce a mixture of polyisobutylene-substituted succinic acylating agents, and

(II) reacting the mixture obtained in step (I) with either (a) an amine having within its structure at least one H-N group, (b) an alcohol, (c) a reactive metal or reactive metal compound, or (d) a mixture of at least two of (a) to (c), the components of (d) being reacted with the acylating agent(s) either simultaneously or sequentially in any order.

The mixture obtained in steps (I) and (II) may thereafter be dissolved in the lubricating oil of the first component containing the polyisobutylene fourth component or may be mixed with the fourth component and the mixture dissolved in the lubricating oil.

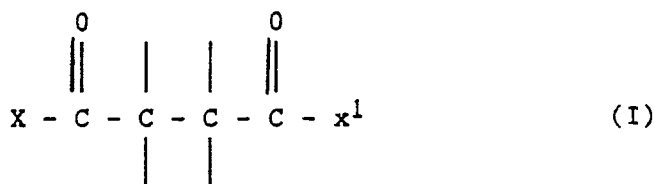
In a modification of the aforesaid method the polyisobutylene having a number average molecular weight in the range from about 40,000 to 400,000 may be incorporated into the composition by mixing with the polyisobutylenes of number average molecular weight in the range 700 to 1475 and 1500 to 5000 respectively, reacting the mixture so-obtained in step (I) with the carboxylic acid to produce a mixture of polyisobutylene-substituted succinic acylating agents and unreacted polyisobutylene having a number average molecular weight in the range about 40,000 to about 400,000 and thereafter reacting with (a) to (d) in step (II). The amount of derivitised polyisobutylene of number average molecular weight in the range from about 40,000 to about 400,000 in the final composition resulting from the modified preparative method will be relatively small because of the relatively small number of reactive sites present in such high molecular weight materials.

The present invention also provides a finished lubricating oil composition comprising a major proportion of an oil of lubricating viscosity and a minor proportion of the concentrate composition as hereinbefore described.

Suitably the amount of the concentrate composition in the finished lubricating oil may be in the range from 0.01 to 10% by weight.

Polyisobutylenes having number average molecular weights in the range from 700 to 1475, 1500 to 5000 and 40,000 to 400,000 are available commercially, for example from BP Chemicals Limited (Hyvis [RTM]), and may be used without further processing.

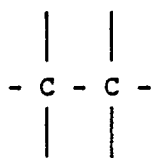
The succinic acylating moiety of the polyisobutylene-substituted succinic acylating agent may suitably have the formula:



wherein X and X¹ are the same or different provided that at least one of X and X¹ is such that the substituted succinic acylating agents can function as carboxylic acylating agents. That is, at least one of X and X¹ must be such that the substituted acylating agent can esterify alcohols, form amides or amine salts with ammonia or amines, form metal salts with reactive metals or basic metal compounds, and otherwise function as a conventional carboxylic acid acylating agent.

Thus X and/or X¹ may be -OH, -O-hydrocarbyl, -O-M⁺ where M⁺ represents one equivalent of a metal, ammonium or amine cation, -NH₂, -Cl, -Br, and together, X and X¹ can be -O- so as to form the anhydride. The specific identity of any X or X¹ group which is not one of the foregoing is not critical so long as its presence does not prevent the remaining group from entering into acylation reactions. Preferably, however, X and X¹ are such that both carboxyl functions of the succinic group can enter into acylation reactions.

One of the unsatisfied valencies in the grouping



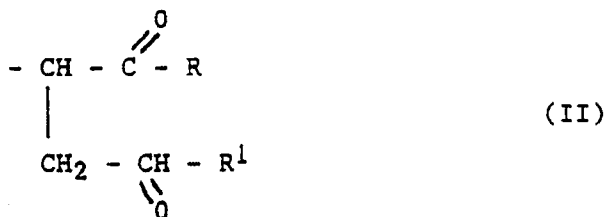
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of the formula (I) forms a carbon to carbon bond with a carbon in the substituent group. While other such unsatisfied valence may be satisfied by a similar bond with the same or different substituent group, all but the one aforesaid valence is usually satisfied by hydrogen.

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The succinic groups will generally correspond to the formula:-

15

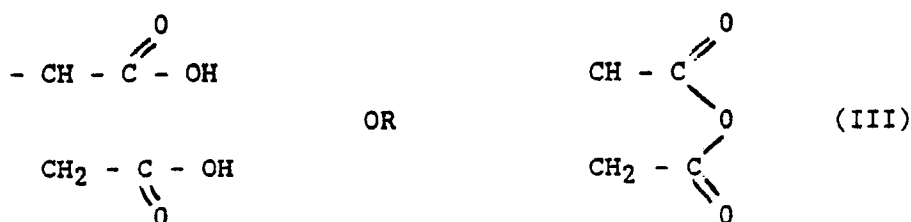


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wherein R and R¹ are each independently either -OH, -Cl, 0-lower alkyl or taken together are -O-. In the latter case the succinic group is a succinic anhydride group. Preferably, the succinic group will corresponds to

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The polyisobutene-substituted succinic acylating agents may be prepared in known manner by reacting a polyisobutylene or mixture as aforesaid with one or more of maleic acid, maleic anhydride, fumaric acid or fumaric anhydride, preferably either maleic acid or maleic anhydride. It is preferred to employ the thermal route in preference to the chlorination route as described in GB-A-1565627 in order to avoid contamination of the product with chlorine.

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The polyisobutylene-substituted succinic acylating agent(s) is then reacted with (a) to (d) in known manner. As regards the amine (a) this may be either a monoamine or a polyamine. Preferably the amine contains at least one primary amino group and more preferably the amine is a polyamine, most preferably a polyamine containing at least two H - N groups, either or both of which are primary or secondary. Suitable amines and polyamines are described in, for example, US-A-3,087,936 and GB-A-1,565,627, the relevant content of which is incorporated by reference herein. Preferred amines (a) include diethylene triamine, triethylene tetramine and tetraethylene pentamine.

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As regards the alcohol (b), this may be either a monohydric or a polyhydric alcohol, polyhydric alcohols being preferred. Suitable alcohols are described in GB-A-1,565,627, the relevant content of which is incorporated herein by reference.

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Reactive metals and reactive metal compounds (c) are those which are known to form salts and complexes when reacted with carboxylic acid and carboxylic acid acylating agents. Further details of suitable compounds are disclosed in GB-A-1,565,627 the relevant parts of which are incorporated by reference herein.

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The oil of lubricating viscosity present in both the concentrate composition and the finished lubricating oil composition may be the same or different. The oil may be a natural oil or a synthetic oil or a mixture of oils. Suitable oils are those described in GB-A-1,565,627, the disclosure of which is incorporated by reference herein.

The finished lubricating oil may contain additional additives conventionally employed in lubricating oils. These may be added directly to the finished lubricating oil composition or may be added in the concentrate composition.

The concentrate composition of the present invention imparts to lubricating oils an increase in their high temperature viscosity characteristics whilst retaining or improving their low temperature viscosity characteristics and providing dispersancy/detergency properties. We have attempted to express the viscosity characteristics by the following relationship:-

$$\text{Numerical gradient} = 100 \times (\text{HTV}_{\text{compound oil}} - \text{HTV}_{\text{oil}}) / (\text{LTV}_{\text{oil}} - \text{HTV}_{\text{oil}})$$

where $\text{HTV}_{\text{compound oil}}$ = High temperature viscosity of the compounded oil.

HTV_{oil} = High temperature viscosity of the lubricating oil.

LTV_{oil} = Low temperature viscosity of the lubricating oil.

Whereas this gradient for the succinimide derivative of either of the second and third components or their combination does not exceed about 11, the gradient for the succinimide derivatives of the second and third components in admixture with the fourth component is about 18.

The invention will now be further illustrated by reference to the following Examples and Comparison Tests.

In the following Examples and Comparison Tests the following commercially available polyisobutenes were employed:-

Commercially Available PIB	No average M.Wt. $\overline{M}_n$	Viscosity average M.Wt. $\overline{M}_v$
(A)	750	-
(B)	810	-
(C)	1000	-
(D)	2400	-
(E)	3800	-
(F)	5800	-
(G)		400,000

Test

Different materials were blended into oil with the aim of improving viscometric performance compared to commercially available materials. The acceptance criterion for improved viscometrics were (at 5% in 150SN lubricating oil):-

V_{100} greater than or equal to 6.9 cSt
and V_{-20} less than or equal to 40P

Comparison Tests 1 - 3

The viscometric characteristics of three commercially available dispersants were determined at 10% in 150 SN lubricating oil. The results are given in Table 1.

Table 1

Material	CT	V ₁₀₀ (cSt)	V ₄₀ (cSt)	V ₋₂₀ (cSt)	VI	Acceptability	
						V ₁₀₀	V ₋₂₀
Dispersant (a) (5.5% actives)	1	8.4	53.2	38	119	✓	✓
Dispersant (b) (7% actives)	2	7.04	45.7	36	112	✓	✓
Dispersant (c)	3	7.07	46.4	37	110	✓	✓

All the commercially available dispersants tested were viscometrically acceptable.

Comparison Tests 4 - 9

The viscometric characteristics of six commercially available polyisobutenes [(A) to (F)] were determined at 5% in 150 SN lubricating oil. The results are given in Table 2.

Table 2

CT	Polyisobutene	V ₁₀₀ (cSt)	V ₄₀ (cSt)	V ₋₂₀ (cSt)	VI	Acceptability	
						V ₁₀₀	V ₋₂₀
4	A	6.1	35.5	31	119	X	✓
5	B	6.1	39	32	101	X	✓
6	C	6.3	38.4	36	113	X	✓
7	D	7.9	52.9	42	116	✓	X
8	E	8.9	59.4	51	126	✓	X
9	F	10.7	68.2	54	146	✓	X

None of the commercially available polyisobutenes tested met the viscometrics acceptance criterion.

Comparison Tests 10 to 17

The viscometric characteristics of eight blends of two commercially available polyisobutenes were determined at 5% in 150 SN lubricating oil. The results are given in Table 3.

Table 23

CT	BLEND		V ₁₀₀ (cSt)	V ₄₀ (cSt)	V ₋₂₀ (cSt)	VI	Acceptability	
	PIB (% w/w)	PIB (% w/w)					V ₁₀₀	V ₋₂₀
10	E (10)	B (90)	6.2	38.6	34	107	X	✓
11	E (20)	B (80)	6.5	41.0	36	109	X	✓
12	E (30)	B (70)	6.9	39.6	37	134	✓	✓
13	F (20)	A (80)	6.8	43.2	35	113	X	✓
14	F (40)	A (60)	7.5	43.4	40	140	✓	✓
15	F (40)	B (60)	7.7	48	40	127	✓	✓
16	F (60)	B (40)	8.6	57	46	125	✓	✓
17	D (60)	C (40)	6.9	44	30	114	✓	✓

Although some of the polyisobutene two-component blends met the viscometric acceptance criterion, the criterion was not consistently met.

Examples 1 to 5

The viscometric characteristics of five three-component polyisobutene blends were determined at 5% in 150 SN lubricating oil. The results are given in Table 4.

Table 4

CT	BLEND		V ₁₀₀ (cSt)	V ₄₀ (cSt)	V ₋₂₀ (cSt)	VI	Acceptability	
	D/C (60:40) (% w/w)	G (% w/w)					V ₁₀₀	V ₋₂₀
	100	0	6.9	44	30	114	✓	✓
1	99	1	7.7	46.7	32	113	✓	✓
2	98	2	7.7	47.7	35	121	✓	✓
3	97	3	7.2	51.2	37	116	✓	✓
4	96	4	8.5	52.5	35	123	✓	✓
5	95	5	8.5	54.5	37	126	✓	✓

The three-component blends consistently meet the viscometric acceptance criterion.

Claims

1. A concentrate composition for use as a multifunctional additive in finished lubricating oils which composition as a first component comprises an oil of lubricating viscosity, as a second component a derivative of a polyisobutylene-substituted succinic acylating agent wherein the polyisobutylene substituent has a number average molecular weight in the range from about 700 to about 1475, as a third component a derivative of a polyisobutylene-substituted succinic acylating agent wherein the polyisobutylene has a number average molecular weight in the range from about 1500 to about 5000 and as a fourth component a polyisobutylene having a number average molecular weight in the range from about 40,000 to about 400,000, the derivatives being formed by reacting the acylating agents either individually or in combination with either (a) an amine having within its structure at least one H-N< group, (b) an alcohol, (c) a reactive metal or reactive metal compound, or (d) a mixture of at least two of (a) to (c), the components of (d) being reacted with the acylating agent(s) either simultaneously or sequentially in any order, the polyisobutylene substituent of the second component forming from 30 to 94.8% by weight of the total polyisobutylene present in the composition, the polyisobutylene substituent of the third component forming from 5 to 69.8% by weight of the total polyisobutylene present in the composition and the polyisobutylene forming the fourth component forming from 0.2 to 5% by weight of the total polyisobutylene present in the composition.

2. A concentrate composition according to claim 1 wherein a mixture of the second and third components is produced by the steps of:-

(I) reacting a mixture of a polyisobutylene having a number average molecular weight in the range from about 700 to about 1475 and a polyisobutylene having a number average molecular weight in the range from about 1500 to about 5000 with an unsaturated carboxylic acid or anhydride thereof to produce a mixture of polyisobutylene-substituted succinic acylating agents, and

(II) reacting the mixture obtained in step (I) with either (a) an amine having within its structure at least one H-N group, (b) an alcohol, (c) a reactive metal or reactive metal compound, or (d) a mixture of at least two of (a) to (c), the components of (d) being reacted with the acylating agent(s) either simultaneously or sequentially in any order.

3. A concentrate composition according to claim 2 wherein the mixture obtained in steps (I) and (II) is dissolved in the oil of lubricating viscosity (the first component) containing the polyisobutylene having a number average molecular weight in the range from about 40,000 to about 400,000 (the fourth component).

4. A concentrate composition according to claim 2 wherein the mixture obtained in steps (I) and (II) is mixed with the polyisobutylene having a number average molecular weight in the range from about 40,000 to about 400,000 (the fourth component) and the mixture so-obtained dissolved in the oil of lubricating viscosity (the first component).

5. A concentrate composition according to claim 2 wherein the polyisobutylene having a number average molecular weight in the range from about 40,000 to 400,000 (the fourth component) is incorporated into the composition by mixing with the polyisobutylenes of number average molecular weight in the range 700 to 1475 and 1500 to 5000 respectively, reacting the mixture so-obtained in step (I) with the carboxylic acid to produce a mixture of polyisobutylene-substituted succinic acylating agents and unreacted polyisobutylene having a number average molecular weight in the range about 40,000 to about 400,000 and thereafter reacting with either (a) to (d) in step (II).

6. A concentrate composition according to any one of the preceding claims wherein the polyisobutylene-substituted succinic acylating agents are prepared by reacting a polyisobutylene or mixture thereof with one or more of maleic acid or maleic anhydride.

7. A concentrate composition according to claim 6 wherein reaction is effected by the thermal route.

8. A concentrate composition according to any one of the preceding claims wherein the derivative is formed by reacting the acylating agent with an amine which is either diethylene triamine, triethylene tetramine or tetraethylene pentamine.

9. A finished lubricating oil composition comprising a major proportion of an oil of lubricating viscosity and a minor proportion of the concentrate composition of claims 1 to 8.

10. A finished lubricating oil composition according to claim 9 wherein the amount of the concentrate composition in the finished composition is in the range from 0.01 to 10% by weight.



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.4)
P,X	EP-A-0 307 132 (EXXON CHEMICAL PATENTS) * Page 5, line 20 - page 6, line 31; page 7, lines 42-46; page 10, lines 6-21; page 13, lines 24-37; page 18, lines 1-29; examples 1-5 *	1,6-10	C 10 M 161/00 // (C 10 M 161/00 C 10 M 129:93 C 10 M 129:95 C 10 M 133:52 C 10 M 143:06) C 10 N 20:04
P,A	---	2-5	
A	GB-A-1 365 311 (OROBIS LTD) * Example A; example 1; claims 1-8 *	1	
A	---		
A	EP-A-0 208 560 (EXXON CHEMICAL PATENTS) * Claims 1-14,17-20,25,26; page 22, lines 9-20 *	1,6-10	
A	---		
A	EP-A-0 264 247 (EXXON CHEMICAL PATENTS) * Page 2, lines 1-11; page 4, lines 19-65; page 5, lines 50-55; page 7, lines 55-65; page 9, lines 27-39; page 10, line 63 - page 11, line 10 *	1,6-10	
A	---		
A	US-A-3 697 428 (N.A. MEINHARDT) * Column 6, example 1; column 7, example 4; column 11, composition D *	1,6,7,9,10	C 10 M
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A	GB-A-2 048 935 (OROBIS LTD) * Page 4, line 45 - page 5, line 4; page 6, example 28; claims 1,6,7,12 *	1,6-10	

The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 28-07-1989	Examiner HILGENGA K.J.
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			