

(12) **EUROPEAN PATENT APPLICATION**

(21) Application number: 81304395.7

(51) Int. Cl.³: **C 10 M 1/18**
C 10 M 1/14, C 10 M 3/08
C 10 M 3/12

(22) Date of filing: 24.09.81

(30) Priority: 24.09.80 GB 8030785

(43) Date of publication of application:
31.03.82 Bulletin 82/13

(84) Designated Contracting States:
AT BE DE FR GB IT NL SE

(71) Applicant: **Orobis Limited**
Belgrave House 76 Buckingham Palace Road
London SW1W 0SU(GB)

(72) Inventor: **Brankling, David**
Orobis Limited Saltend
Hedon Hull, HU12 8DS(GB)

(72) Inventor: **Crawford, John**
Orobis Limited 36 - 44 High Street
Redhill Surrey, RHL 1RW(GB)

(74) Representative: **Harry, John**
BP INTERNATIONAL LIMITED Patents and Licensing
Division Chertsey Road
Sunbury-on-Thames Middlesex TW16 7LN(GB)

(54) Viscosity index improver additive composition.

(57) A viscosity index improver additive composition comprising from 1 to 25% by weight of a hydrogenated alkenylarene/conjugated diene interpolymers having a number average molecular weight greater than 20,000, from 30 to 72% by weight of a synthetic ester lubricating oil comprising an alkyl ester of a saturated aliphatic carboxylic acid and from 15 to 50% by weight of a non-synthetic mineral lubricating oil, all percentages by weight being expressed in terms of the total weight of the composition. In a modification of the invention the proportion of mineral lubricating oil is decreased and the proportion of ester lubricating oil is increased by the addition of, for example, either an ethylene copolymer or a polybutene in an amount of from 1 to 25% by weight.

VISCOSITY INDEX IMPROVER ADDITIVE COMPOSITION.

The present invention relates to a viscosity index improver additive composition and to a finished lubricating oil containing the additive composition.

Lubricating oils are normally classified in terms of their
5 viscosity at some standard temperature but equally important is a property known as the viscosity index, which is an empirical number giving a measure of the extent to which the viscosity of an oil decreases as the temperature is raised. An oil which satisfies viscosity requirements at both extremes of the temperature range
10 to which it may be subjected is said to have a high "viscosity index". This property can be controlled to some extent by refining, but in recent years the trend has been towards 'multi-grade' oils, of extremely high viscosity index, in which certain polymer compounds which function as viscosity index improvers are added. In
15 addition to the properties of improving viscosity index and of being stable under conditions of high shear, it is necessary for any potential lubricating oil additives to have two other important properties, namely compatibility with the lubricating oil and the stability under conditions of oxidation which might reasonably be
20 expected to be encountered under conditions of storage and use of such compositions.

Recently, there has been developed a series of effective viscosity index improving agents for lubricating compositions based upon various alkenylarene/conjugated diene interpolymers which are
25 generally hydrogenated to improve their oxidative stability. These

inter polymer additives are based upon several types of alkenylarene/
conjugated diene copolymers which differ from each other, principally,
in the steric arrangement of the polymerised monomers. On the one
hand US Patent Nos: 3,554,911 (Schiff et al), 3630905 (Sorgo),
5 USP 3752767 (Eckert) and 3772169 (Small et al), to select but a few,
are concerned with the use of hydrogenated random butadiene/styrene
copolymers as VI improvers for lubricating oils. These copolymers are
prepared by the copolymerisation, using conventional techniques, of
butadiene and styrene in the presence of a randomising agent and
10 subsequently the copolymers are partially hydrogenated. On the other
hand US Patent Nos: 3994815 (Coleman), 3775329 (Eckert), 3668125
(Anderson) and 3763044 (Anderson), for example are concerned with the
use of hydrogenated block and tapered copolymers of an alkenylarene,
e.g. styrene or alphas-methylstyrene and a conjugated diene, e.g.
15 butadiene or isoprene as viscosity index improver additives. These
polymers are composed of essentially pure blocks of the individual
polymers and are prepared by anionic polymerisation techniques,
followed in most cases by hydrogenation.

The difficulties encountered during the formulation of certain
20 lubricating compositions containing these copolymers have been
extensively enumerated in for example US Patent Nos: 3630905 (Sorgo),
3772169 (Small et al) and 3668125 (Anderson) and the complete
specification of UK Patent No: 1540292 (Lubrizol). All these specifi-
cations suggest solutions to these problems. The Sorgo specification
25 discloses the preparation of an oil-extended composition comprising
(a) 40 - 60 weight percent of the particular hydrogenated copolymer
and (b) 60 - 40 weight percent of a paraffin oil. Large amounts of
this oil-extended composition are prepared in one operation and
smaller amounts, as needed, are used in the preparation of the final
30 lubricating compositions. The Small et al patent is concerned with
the prevention of a gelling tendency that mineral lubricating oil
or ester type oil compositions containing these hydrogenated co-
polymers have. Their proposed solution is the addition of small
amounts of a polyester of anolefinically unsaturated acid to the oil
35 solution. GB 1540292 teaches that many of the difficulties encountered

in formulating lubricating compositions containing hydrogenated
interpolymers may be eliminated or diminished by first preparing
an additive concentrate using a synthetic substantially hydrocarbon
alkylated-aromatic lubricating oil diluent or carrier. Optionally
5 supplemental diluents selected from ester synthetic lubricating
oils, mineral lubricating oils and mixtures thereof may be employed.

We have found that the compatibility of hydrogenated alkenyl-
arene/conjugated diene interpolymers in synthetic ester lubricating
oils can be considerably improved by the addition of a mineral
10 lubricating oil.

Accordingly the present invention provides a viscosity index
improver additive composition which composition comprises from
1 to 25% by weight of a hydrogenated alkenylarene/conjugated diene
interpolymer having a number average molecular weight greater than
15 20,000, from 30 to 72% by weight of a synthetic ester lubricating
oil comprising an alkyl ester of a saturated aliphatic carboxylic
acid and from 15 to 50% by weight of a non-synthetic mineral lubricating
oil, all percentages by weight being expressed in terms of the total
weight of the composition.

20 With regard to the hydrogenated alkenylarene/conjugated diene
interpolymer the alkenylarene may be a vinyl mono-, di- or
polyaromatic compound such as styrene or vinyl naphthalene,
preferably styrene, alkylated styrene or halogen-substituted styrene,
and the conjugated diene may be butadiene or isoprene. The inter-
25 polymer may be a random, block, or star-shaped copolymer.
All these terms are well-known in the art and include such polymers
as are described in the aforesaid patent specifications which are
herein incorporated by reference.

Suitable methods of hydrogenation are described for example in
30 US Patent Nos: 3113986, 3205278 and 2864809. The object of hydro-
genation is to saturate the olefinic double bonds in preference to
the aromatic nucleus double bonds. Preferably more than 95% of the
olefinic double bonds and less than 5% of the aromatic nucleus
double bonds are hydrogenated. Suitably the interpolymer has a
35 number average molecular weight in the range 20,000 to 125,000,

preferably from 25,000 to 100,000. A particularly suitable co-polymer is a hydrogenated styrene/isoprene block co-polymer of number average molecular weight in the range 50,000 to 100,000 and containing about 75% isoprene and 25% styrene, greater than 95% of the isoprene component being present in the 1,4- form in which greater than 95% of the olefinic double bonds are hydrogenated and the styrene component having less than 5% of the aromatic nucleus double bonds hydrogenated. Preferably the vinylarene/conjugated diene interpolymers form from 2 to 15% by weight of the composition.

10 The alkyl ester of a saturated aliphatic carboxylic acid may suitably be a simple ester, diester, partial ester or complex ester formed from one or more C_{10} to C_{18} saturated organic mono- or polycarboxylic acids and one or more C_3 to C_9 mono- or polyhydric alcohols. Preferably the ester is formed from two molecular proportions of a monohydric alcohol and one molecular proportion of a dicarboxylic acid, e.g. di(octyl) dodecane dioate. Preferably the synthetic ester lubricant is present in an amount in the range from 50 to 72% by weight.

20 The non-synthetic mineral lubricating oil may suitably be the common solvent-treated or acid-treated mineral oils of the paraffinic, naphthenic or mixed paraffinic-naphthenic types. Preferred oils are the Solvent Neutral oils and in particular 150 Solvent Neutral oil. Preferably the composition contains from 10 to 40% by weight of the non-synthetic mineral lubricating oil.

25 A particularly preferred composition comprises 4.5% by weight of a hydrogenated styrene/isoprene block copolymer of number average molecular weight in the range 50,000 to 100,000 and containing about 75% isoprene and 25% styrene, greater than 95% of the isoprene component being present in the 1,4- form in which greater than 95% of the olefinic double bonds are hydrogenated and the styrene component having less than 5% of the aromatic nucleus double bonds hydrogenated, hereinafter referred to as 'hydrogenated styrene/isoprene tapered' (HSIT) interpolymers, 70% by weight di(octyl) dodecane dioate and 25.5% by weight 150 Solvent Neutral lubricating oil.

35 It has also been unexpectedly found that the proportion of

non-synthetic mineral lubricating oil can be decreased and the proportion of synthetic ester lubricating oil increased by incorporating a small proportion of an olefinic homo- or copolymer such as an ethylene/propylene/non-conjugated diolefin terpolymer or a polybutene, particularly a polybutene, in the composition.

Accordingly the invention also provides a viscosity index improver additive composition as hereinbefore described modified in the respects that the amount of synthetic ester lubricating oil is in the range from 40 to 80% by weight, the amount of non-synthetic mineral lubricating oil is in the range from 15 to 40% by weight and there is included from 1 to 25% by weight of an olefinic polymer.

The olefinic polymer may be a homopolymer or a copolymer. Preferably the olefinic polymer is a polybutene of which polyisobutene is preferred. Suitable polybutenes may have a number average molecular weight in the range from 100 to 50,000.

According to another aspect of the present invention there is provided a finished lubricating oil composition comprising a major proportion of a lubricating oil base and a minor proportion of the viscosity index improver additive composition as described hereinbefore.

The lubricating oil base may be derived from a variety of sources. Thus the base oil may be derived from natural or synthetic sources which include animal oils, vegetable oils, mineral oils, synthetic hydrocarbon oils and synthetic ester oils.

The amount of the additive composition present in the finished lubricating oil composition is an amount sufficient to improve the viscosity index of the composition. Generally from 0.05 to 10% by weight of the additive composition may be used, though in certain specialised applications up to 15% by weight may be required.

In addition to the viscosity index improver additive composition the finished lubricating composition may also contain other additives conventionally employed in the art, such as dispersants, detergents, corrosion inhibitors, anti-wear agents, etc.

The invention will now be illustrated by reference to the following Examples.

Example 1

A composition composed of 10% by weight of a commercially available HSIT interpolpolymer, 60% by weight di(octyl) dodecane dioate and 30% by weight 150 Solvent Neutral oil was found to be compatible.

5 Comparison Test 1

The same commercially available HSIT as used in Example 1 was found to be incompatible with di(octyl) dodecane dioate at all concentrations.

Comparison Test 2

10 A composition composed of 5.6% by weight of the HSIT as used in Example 1, 85% by weight di(octyl) dodecane dioate and 9.4% by weight Solvent Neutral oil resulted in separation i.e. it formed incompatible composition.

Comparison Test 3

15 A composition composed of 5.0% by weight of the HSIT as used in Example 1, 75% by weight of di(octyl) dodecane dioate and 20% by weight Solvent Neutral oil was incompatible.

Example 2

20 A composition containing 5.0% by weight of the HSIT as used in Example 1, 75.0% by weight of di(octyl) dodecane dioate, 2.0% by weight of a solution of a polybutene having a number average molecular weight in the range 30,000 to 42,000 in 150 Solvent Neutral base oil, the polybutene forming 45% by weight of the solution and 18.0% by weight 150 Solvent Neutral base oil was compatible.

25 Comparison Test 4

5% by weight of a commercially available random styrene/butadiene interpolpolymer having a number average molecular weight greater than 20,000 in di(octyl) dodecane dioate gave a cloudy solution, i.e. the composition was incompatible.

30 Example 3

20% by weight of the same styrene/butadiene interpolpolymer as was used in Comparison Test 4, 60% by weight di(octyl) dodecane dioate and 20% 100 Solvent Neutral base oil were mixed to form a compatible solution.

Example 4

A composition composed of 6.85% by weight of the commercially available HSIT interpolymers as used in Example 1, 57.15% by weight of di(octyl) dodecane dioate, 18% by weight of polyalpha olefin (6 cSt) and 18% by weight 150 Solvent Neutral base oil was compatible at room temperature.

10

15

20

25

30

35

Claims:

1. A viscosity index improver additive composition which composition comprises from 1 to 25% by weight of a hydrogenated alkenylarene/ conjugated diene interpolpolymer having a number average molecular weight greater than 20,000, from 30 to 72% by weight of a synthetic ester lubricating oil comprising an alkyl ester of a saturated aliphatic carboxylic acid and from 15 to 50% by weight of a non-synthetic mineral lubricating oil, all percentages by weight being expressed in terms of the total weight of the composition.
2. A composition according to claim 1 wherein the alkenylarene is styrene, and the conjugated diene is either butadiene or isoprene.
3. A composition according to any one of the preceding claims wherein the interpolpolymer is a hydrogenated styrene/isoprene block copolymer of number average molecular weight in the range 50,000 to 100,000 containing about 75% isoprene and 25% styrene, greater than 95% of the isoprene component being present in the 1,4-form in which greater than 95% of the olefinic double bonds are hydrogenated and in which less than 5% of the aromatic nucleus double bonds in the styrene component are hydrogenated.
4. A composition according to any one of the preceding claims wherein the interpolpolymer forms from 2 to 15% by weight of the composition.
5. A composition according to any one of the preceding claims wherein the alkyl ester is a simple ester, a diester, a partial ester or a complex ester formed from one or more C₁₀ to C₁₈ saturated organic mono- or poly-carboxylic acids and one or more C₃ to C₉ mono- or polyhydric alcohols.

6. A composition according to claim 5 wherein the ester is formed from two molecular proportions of a monohydric alcohol and one molecular proportion of a dicarboxylic acid.
7. A composition according to any of the preceding claims wherein the synthetic ester is present in an amount in the range from 50 to 72% by weight.
8. A composition according to any one of the preceding claims wherein the non-synthetic mineral lubricating oil is a Solvent Neutral oil.
9. A composition according to any one of the preceding claims wherein the non-synthetic mineral lubricating oil forms from 10 to 40% by weight of the composition.
10. A composition according to any one of the preceding claims comprising 4.5% by weight of a hydrogenated styrene/isoprene block copolymer of number average molecular weight in the range 50,000 to 10,000 and containing about 75% isoprene and 25% styrene, greater than 96% of the isoprene component being present in the 1,4-form in which greater than 95% of the olefinic double bonds are hydrogenated and the styrene component having less than 5% of the aromatic nucleus double bonds hydrogenated, 70% by weight di(octyl) dodecane dioate and 25.5% by weight 150 Solvent Neutral lubricating oil.
11. A viscosity index improver additive composition as claimed in any one of the preceding claims modified in the respects that the amount of synthetic ester lubricating oil is in the range from 40 to 80% by weight, the amount of non-synthetic mineral lubricating oil is in the range from 15 to 40% by weight and there is included from 1 to 25% by weight of an olefinic polymer.
12. A composition according to claim 11 wherein the olefinic polymer is a polybutene.
13. A finished lubricating oil composition comprising a major proportion of a lubricating oil base and a minor proportion of the viscosity index improver additive composition as claimed in any one of the preceding claims.
14. A finished lubricating oil composition according to claim 13 wherein the additive is present in an amount in the range from 0.05 to 15% by weight.



European Patent
Office

EUROPEAN SEARCH REPORT

0048631

Application number
EP 81 30 4395

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. ³)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
D	<u>GB - A - 1 540 292 (THE LUBRIZOL)</u> * claims 1-7; page 1, line 3 to page 4, line 122; page 5, line 77 to page 6, line 74; page 7, lines 74-90 * & FR - A - 2 298 597 & DE - A - 2 602 342 & US - A - 3 994 815 ---	1-14	C 10 M 1/18 1/14 3/08 3/12
A	<u>FR - A - 2 223 450 (RHONE-PROGIL)</u> * claims 1-6 & DE - A - 2 414 872 & US - A - 3 948 843 & GB - A - 1 445 051 ---	1-4, 11-14	TECHNICAL FIELDS SEARCHED (Int. Cl. ³) C 10 M 1/14 1/18 3/08 3/12
A/D	<u>US - A - 3 772 169 (N.J.H. SMALL et al.)</u> & FR - A - 2 128 590 & DE - A - 2 210 430 & NL - A - 72 02749 ---		
A/D	<u>US - A - 3 630 905 (P.M. SORGO)</u> -----		
			CATEGORY OF CITED DOCUMENTS X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons &: member of the same patent family, corresponding document
☒ The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
The Hague	15.12.1981	RO TSAERT	