



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification⁴ : C10M 141/10,173/02 // (C10M 141/10,133:52, 133:56,137:08) (C10M 173/02,133:52,133:56, 137:08) C10N 60:14	A3	(11) International Publication Number: WO 87/ 07637 (43) International Publication Date: 17 December 1987 (17.12.87)
(21) International Application Number: PCT/US87/01311 (22) International Filing Date: 5 June 1987 (05.06.87) (31) Priority Application Number: 874,267 (32) Priority Date: 13 June 1986 (13.06.86) (33) Priority Country: US (71) Applicant: THE LUBRIZOL CORPORATION [US/US]; 29400 Lakeland Boulevard, Wickliffe, OH 44092 (US). (72) Inventors: SCHWIND, James, J. ; 266 East 323rd Street, Willówick, OH 44094 (US). DI BIASE, Stephen, A. ; 504 East 266th Street, Euclid, OH 44132 (US). (74) Agents: CORDEK, James, L. et al.; The Lubrizol Corporation, 29400 Lakeland Boulevard, Wickliffe, OH 44092 (US).	(81) Designated States: AT (European patent), AU, BE (European patent), BR, CH (European patent), DE (European patent), DK, FI, FR (European patent), GB (European patent), IT (European patent), JP, LU (European patent), NL (European patent), NO, SE (European patent). (88) Date of publication of the international search report: 10 March 1988 (10.03.88) Published <i>With international search report</i> <i>With amended claims.</i> Date of publication of the amended claims: 10 March 1988 (10.03.88)	
(54) Title: PHOSPHORUS-CONTAINING LUBRICANT AND FUNCTIONAL FLUID COMPOSITIONS		
<div style="text-align: center; margin: 20px 0;"> $\begin{array}{l} R^1O \\ \diagdown \\ P(X)XH \\ \diagup \\ R^2O \end{array} \quad (I)$ </div>		
(57) Abstract <p>Lubricating oil and functional fluid compositions having improved high temperature stability and which contain at least one phosphorus-containing composition and at least one oil-soluble nitrogen-containing composition. More particularly, the lubricating and functional fluid compositions of the present invention comprise (A) a major amount of an oil of lubricating viscosity, and a minor amount of (B-1) at least one soluble amine salt of at least one substituted phosphoric acid composition characterized by formula (I), wherein R¹ is hydrogen or a hydrocarbyl group, R² is a hydrocarbyl group, and both X groups are either O or S, and (C) at least one soluble nitrogen-containing composition prepared by the reaction of a hydrocarbon-substituted succinic acid-producing compound with at least about one-half equivalent, per equivalent of acid producing compound, of an amine containing at least one hydrogen attached to a nitrogen atom. Preferably, the amine salts of the phosphoric acids utilized in the lubricating compositions of the present invention are derived from primary amines, and the soluble nitrogen-containing compositions (C) also contain boron. The lubricating compositions of the present invention are particularly useful in gear applications requiring high thermal stability such as from about 160°C with intermittent operation up to about 200°C.</p>		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FR	France	ML	Mali
AU	Australia	GA	Gabon	MR	Mauritania
BB	Barbados	GB	United Kingdom	MW	Malawi
BE	Belgium	HU	Hungary	NL	Netherlands
BG	Bulgaria	IT	Italy	NO	Norway
BJ	Benin	JP	Japan	RO	Romania
BR	Brazil	KP	Democratic People's Republic of Korea	SD	Sudan
CF	Central African Republic	KR	Republic of Korea	SE	Sweden
CG	Congo	LI	Liechtenstein	SN	Senegal
CH	Switzerland	LK	Sri Lanka	SU	Soviet Union
CM	Cameroon	LU	Luxembourg	TD	Chad
DE	Germany, Federal Republic of	MC	Monaco	TG	Togo
DK	Denmark	MG	Madagascar	US	United States of America
FI	Finland				

AMENDED CLAIMS

[received by the International Bureau on 9 February 1988 (09.02.88);
original claims 1-63 replaced by new claims 1-17 (5 pages)]

1. A lubricating or functional fluid composition having improved high temperature stability comprising

(A) a major amount of an oil of lubricating viscosity, and a minor amount of

(B-1) at least one soluble tertiary aliphatic primary amine salt wherein the primary amine contains from about 4 to about 30 carbon atoms of at least one substituted phosphoric acid composition characterized by the formula



wherein R^1 is hydrogen or an aliphatic hydrocarbyl group, R^2 is an aliphatic hydrocarbyl group wherein R^1 and R^2 contain from about 4 to about 60 carbon atoms, and both X groups are either O or S, and

(C) at least one soluble nitrogen- and boron-containing compound prepared by the reaction of

(C-1) at least one boron compound selected from the class consisting of boron trioxide, boron halides, boron acids, boron anhydrides, boron amides and esters of boron acids with

(C-2) at least one soluble acylated nitrogen intermediate prepared by the reaction of a hydrocarbon-substituted succinic acid-producing compound selected from the group consisting of succinic acids, anhydrides, ester and halides containing an average of at least about 50 aliphatic carbon atoms in the substituent with at least about one-half equivalent, per equivalent of acid producing compound, of an amine containing at least one hydrogen attached to a nitrogen atom.

2. The composition of claim 1 wherein the phosphoric acid composition of (B-1) comprises a mixture of substituted phosphoric acids prepared by the reaction of at least one hydroxy compound with a phosphorus reactant of the formula P_2X_5 wherein X is O.

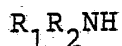
3. The composition of claim 1 also containing at least one (B-2) di-hydrocarbyl-substituted phosphite characterized by the formula



wherein each R is a hydrocarbyl group which may be the same or different.

4. The composition of claim 1 wherein the hydrocarbon substituent of the succinic acid-producing compound of (C-2) is derived from a polyolefin having an Mn value within the range of from about 700 to about 10,000.

5. The composition of claim 1 wherein the amine of (C-2) is characterized by the formula



wherein R_1 and R_2 are each independently hydrogen, or hydrocarbon, amino-substituted hydrocarbon, hydroxy-substituted hydrocarbon, alkoxy-substituted hydrocarbon, amino, carbamyl, thiocarbamyl, guanlyl, and acylimidoyl groups, provided that only one of R_1 and R_2 may be hydrogen.

6. The composition of claim 1 wherein the amine of (C-2) is a polyamine.

7. The composition of claim 1 wherein the amine of (C-2) is a hydroxyalkyl-substituted alkylene polyamine.

8. The composition of claim 1 wherein the boron compound is boric acid.

9. The composition of claim 1 wherein the weight ratio of (B-1):(C) is from about 0.1:1 to about 10:1.

10. The composition of claim 1 wherein the amount of (C-1) and (C-2) present is an amount to provide from about 0.1 atomic proportion of boron for each mole of said acylated nitrogen intermediate to about 10 atomic proportions of boron for each atomic proportion of nitrogen of said acylated nitrogen intermediate and wherein the soluble nitrogen- and boron-containing composition (C) is prepared by reacting (C-1) with (C-2) at an elevated temperature up to, but not including the decomposition temperature of any reactants or the product of the reaction.

11. A lubricating or functional fluid composition having improved high temperature stability comprising

(A) a major amount of an oil of lubricating viscosity, and a minor amount of

(B-1) at least one soluble amine salt of a tertiary aliphatic primary amine containing from about 4 to about 30 carbon atoms with a substituted phosphoric acid composition characterized by the formula



wherein R^1 and R^2 are aliphatic hydrocarbyl groups and together contain from about 4 to about 60 carbon atoms, and both X groups are either O or S, and

(C) at least one soluble nitrogen- and boron-containing composition prepared by the reaction of

(C-1) at least one boron compound selected from the class consisting of boron trioxide, boron halides, boron acids, boron anhydrides, boron amides and esters of boron acids with

(C-2) at least one soluble acylated nitrogen intermediate prepared by the reaction of an aliphatic olefin polymer-substituted succinic acid-producing compound having an average of at least about 50 aliphatic carbon atoms in the polymer substituent with at least about one-half equivalent, for each equivalent of acid-producing compound, of an alkylene amine, a hydroxyalkyl-substituted alkylene amine, or a mixture thereof.

12. The composition of claim 11 wherein the phosphoric acid composition of (B-1) comprises a mixture of substituted phosphoric acids characterized by the formula



wherein R^1 is hydrogen or a hydrocarbon group and R^2 is a hydrocarbon group, the total number of carbon atoms in R^1 and R^2 being from about 4 to about 60.

13. The composition of claim 11 also containing (B-2) at least one di-hydrocarbyl-substituted phosphite characterized by the formula



wherein each R is a hydrocarbyl group which may be the same or different.

14. The composition of claim 11 wherein the polymer substituent of the succinic acid-producing compound is derived from a polybutene having an Mn value within the range of from about 700 to about 10,000.

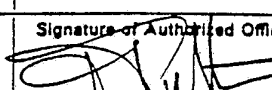
15. The composition of claim 11 wherein the weight ratio of (B-1):(C) is from about 0.1:1 to about 10:1.

16. The composition of claim 11 wherein the amount of (C-1) and (C-2) present is an amount sufficient to provide from about 0.1 atomic proportion of boron for each mole of said acylated nitrogen intermediate up to about 10 atomic proportions of boron for each atomic proportion of nitrogen of said acylated nitrogen intermediate and wherein the soluble nitrogen- and boron-containing composition (C) is prepared by reacting (C-1) with (C-2) at a temperature of from about 50°C to about 250°C.

17. The composition of claim 11 containing from about 0.1% to about 5% by weight of (B) and from about 0.1% to about 5% by weight of (C).

INTERNATIONAL SEARCH REPORT

International Application No PCT/US 87/01311

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶		
According to International Patent Classification (IPC) or to both National Classification and IPC		
IPC ⁴ : C 10 M 141/10; 173/02 // (C 10 M 141/10, 133:52, 133:56, 137:08)(C 10 M 173/02, 133:52, 133:56, 137:08) C 10 N 60:		
II. FIELDS SEARCHED		14
Minimum Documentation Searched ⁷		
Classification System	Classification Symbols	
IPC ⁴	C 10 M	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸		
III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹		
Category ⁹	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
X	US, A, 3267033 (J.W. ALLEN) 16 August 1966 see claims 1-4; column 1, line 6 - column 3, line 35; column 5, lines 12-14; column 7, lines 25-30 --	1-11,13-18
X	US, A, 3901932 (FUSAO TADA et al.) 26 August 1975 see column 14, lines 3-25; column 15, lines 5-25; table 5, examples r,s --	1-11,13-58
X	US, A, 4472288 (K.A. FROST) 18 September 1984 see claims 1,7-10; column 4, lines 24,25 --	1-11,13-58
X	GB, A, 1117349 (SHELL) 19 June 1968 see claims 1-10 --	1-11,13-18
X	US, A, 4163729 (J.H. ADAMS) 7 August 1979 see claim 1; column 3, lines 58-60; column 8, line 48 - column 10, line 3 --	1-4,6-11,13-18 ./.
<p>⁹ Special categories of cited documents: ¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p>		
IV. CERTIFICATION		
Date of the Actual Completion of the international Search		Date of Mailing of this International Search Report
8th December 1987		20 JAN 1988
International Searching Authority		Signature of Authorized Officer
EUROPEAN PATENT OFFICE		 P.C.G. VAN DER PUTTEN

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category*	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
X	US, A, 4431552 (C.G. SALENTINE) 14 February 1984 see claims 1,2,5-7; column 2, line 40 - column 3, line 24; column 3, lines 53-54	1-11,13-18
	--	
X	DE, A, 2823877 (EXXON) 13 December 1979 see claims 1,2,5-9; page 14, paragraph 3	1-11,13-18
	--	
X	GB, A, 2052505 (LUBRIZOL) 28 January 1983 see claim 1; page 1, lines 52-55; page 4, lines 39-55; page 5, lines 8-20	12,30,47-49
	--	
X	FR, A, 1367789 (LUBRIZOL) 1964 see claim A1°; page 3, column 1, paragraphs 2,3; page 3, column 2, paragraph 4 - page 4, column 2, paragraph 2; page 10, column 1, lines 1-5	1-11,13-18
	--	
X	US, A, 4337161 (R.A. STAYNER) 29 June 1982 see claim 1; column 2, lines 26-28, 57-59; column 3, lines 6-14; column 3, lines 44-52	61

**ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO.**

US 8701311

SA 17944

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on 22/12/87. The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A- 3267033		None	
US-A- 3901932	26-08-75	GB-A- 1420543 JP-A- 49126623	07-01-76 04-12-74
US-A- 4472288	18-09-84	None	
GB-A- 1117349		NL-A- 6609187 DE-A- 1645883 FR-A- 1525398	16-10-67 02-01-70
US-A- 4163729	07-08-79	US-A- 4089790	16-05-78
US-A- 4431552	14-02-84	None	
DE-A- 2823877	13-12-79	AU-A- 3634778 AU-B- 515964	29-11-79 14-05-81
GB-A- 2052505	28-01-81	FR-A, B 2458555 JP-A- 56002397 US-A- 4263150 CA-A- 1134851	02-01-81 12-01-81 21-04-81 02-11-82
FR-A- 1367789		DE-A- 1254146 GB-A- 1009914 US-A- 3197405	
US-A- 4337161	29-06-82	None	