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(54) **LUBRICATING OIL ADDITIVE AND LUBRICATING OIL COMPOSITION**

(57) The present invention is directed to a lubricating oil additive containing a reaction product of a succinimide compound and a phosphorus atom-containing compound, and to a lubricating oil composition containing a

mineral oil and/or a synthetic base oil, and the above lubricating oil additive.

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DescriptionTechnical Field

5 **[0001]** The present invention relates to a lubricating oil additive and to a lubricating oil composition. More specifically, the present invention is directed to a lubricating oil additive which can afford a lubricating oil composition, such as an automatic transmission fluid or a continuously variable transmission fluid, having a particularly high wet friction material torque capacity and excellent μ (friction coefficient)-V (sliding velocity) performances, and to a lubricating oil composition containing such a lubricating oil additive and having the above properties.

Background Art

[0002] In recent years, there has been an increasing demand for a reduction of fuel consumption of automobiles for the purpose of reducing the discharge amount of carbon dioxide in view of the global environmental problems.

15 **[0003]** In particular, in transmissions, an improvement of power transmission efficiency is going to be more demanded than ever and, thus, a high torque capacity is required for a lubricating oil which is one of the important constituent components thereof.

[0004] On the other hand, as an effective means to improve the power transmission efficiency, use has been recently made of a lock-up clutch, which is effective in improving the fuel consumption, in automatic transmission and continuously variable transmission of automobiles.

[0005] In this mechanism, the transmission is built in a torque converter.

[0006] Such a lock-up clutch has a function to transmit the driving power from an engine directly to the transmission according to the running conditions. By shifting between the torque converter drive and the direct drive at proper timings, the efficiency of the torque converter can be improved.

25 **[0007]** Hitherto, use has been proposed of a phosphate ester, a fatty acid ester, a fatty amide or the like as a friction modifier for an automatic transmission fluid or a continuously variable transmission fluid.

[0008] The incorporation of such a friction modifier, however, has a defect that the friction coefficient in a low velocity slip area of the lock-up clutch part is reduced and the friction coefficient of a wet clutch is considerably reduced, so that a sufficient transmission torque capacity cannot be ensured.

30 **[0009]** For this reason, a metal-type detergent such as calcium sulfonate has been incorporated for the purpose of improving the transmission torque capacity.

[0010] The addition of these substances, however, causes a problem that the friction material is clogged upon use for a long period of time so that the friction coefficient is reduced and the friction characteristics, such as μ -V performances of the lock-up clutch part, are deteriorated.

35 **[0011]** To cope with this problem, a lubricating oil composition containing a bisimide and a monoimide having a hydrocarbyl group with 8 to 30 carbon atoms is disclosed (for example, see JP-A-2002-105478) as a composition exhibiting both high torque capacity of a wet clutch and good transmission performances while maintaining the shudder vibration preventing performance in a low speed range.

40 **[0012]** A lubricating oil composition containing a bisimide compound having a hydrocarbyl group with 8 to 30 carbon atoms and a boron-modified ashless dispersant is disclosed (for example, see JP-A-2001-288489) as a composition capable of maintaining shudder vibration preventing performance in a low speed range and of preventing delamination in the clutch.

[0013] With such lubricating oil compositions, however, a problem has been pointed out that a sufficient torque capacity cannot be ensured.

45 **[0014]** With the foregoing background in view, there is a demand, in the field of automatic transmission fluids and continuously variable transmission fluids, for the development of a technique for satisfying required friction performances of a wet friction material, particularly a technique for increasing the friction coefficient of a wet clutch, while maintaining freedom of metal components or maintaining a metal content at a low level.

50 **[0015]** The present invention has been made in the above situation and has as its object the provision of a lubricating oil additive which can afford a lubricating oil composition, such as an automatic transmission fluid or a continuously variable transmission fluid, having a particularly high wet friction material torque capacity and excellent μ -V performances, and of a lubricating oil composition containing such a lubricating oil additive and having the above properties.

Disclosure of the Invention

55 **[0016]** The present inventors have made an intensive study to achieve the foregoing objects and, as a result, have found that a reaction product of a succinimide compound and a phosphorus atom-containing compound can serve as a lubricating oil additive meeting with the objects.

[0017] The present invention has been completed on the basis of such a finding.

[0018] Thus, the present invention provides:

- (1) a lubricating oil additive comprising a reaction product of a succinimide compound and a phosphorus atom-containing compound;
- (2) a lubricating oil additive of (1) above, wherein the phosphorus atom-containing compound is at least one compound selected from the group consisting of phosphorus sulfide, a phospho sulfurized hydrocarbon compound, a phosphate ester, a phosphite ester and a dithiophosphate ester;
- (3) a lubricating oil additive of (2) above, wherein the phospho sulfurized hydrocarbon compound has a structure in which two alkyl groups are bonded to a phosphorus atom;
- (4) a lubricating oil additive of (2) above, wherein the phosphate ester, phosphite ester and dithiophosphate ester are each a diester;
- (5) a lubricating oil additive of any one of (2) to (4) above, wherein the alkyl group introduced into the phosphorus atom-containing compound is an alkyl group which has 2 to 25 carbon atoms and which may have an ether bond and/or a thioether bond;
- (6) a lubricating oil composition characterized by comprising a mineral oil and/or a synthetic base oil, and a lubricating oil additive according to any one of (1) to (5) above;
- (7) a lubricating oil composition of (6) above, wherein the composition is used for a transmission having a wet clutch or a wet brake; and
- (8) a lubricating oil composition of (6) above, wherein the composition is an automatic transmission fluid or a continuously variable transmission fluid.

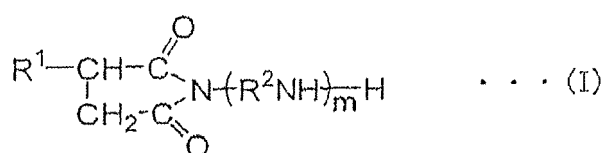
[0019] According to the present invention, there can be provided a lubricating oil additive which can afford a lubricating oil composition, such as an automatic transmission fluid or a continuously variable transmission fluid, having a particularly high wet friction material torque capacity and excellent μ -V performances, and a lubricating oil composition containing such a lubricating oil additive and having the above properties.

Best Mode for Carrying Out the Invention

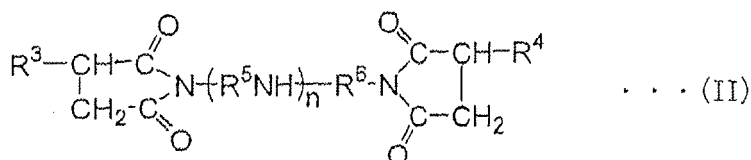
[0020] The lubricating oil additive of the present invention is a reaction product of (A) a succinimide compound and (B) a phosphorus atom-containing compound.

[0021] In the lubricating oil additive of the present invention, the succinimide compound used as the raw material (A) may be a boron-free succinimide compound or a boron-containing succinimide compound.

[0022] The boron-free succinimide compound may be, for example, a succinimide represented by the general formula (I):



or by the general formula (II):



[0023] In the general formulas (I) and (II), R^1 and R^3 each represent an alkyl group or an alkenyl group having 5 to 350 carbon atoms and R^4 represents hydrogen, or an alkyl group or an alkenyl group having 5 to 350 carbon atoms.

[0024] R^3 and R^4 may be the same with or different from each other.

[0025] R^2 , R^5 and R^6 each represent a divalent organic group and R^5 and R^6 may be the same with or different from each other.

[0026] Represented by m is an integer of 1 to 10 and by n is an integer of 1 to 10.

[0027] The above alkyl or alkenyl group is preferably a polyalkenyl group, particularly a polybutenyl group or a polyisobutenyl group.

[0028] The succinimide represented by the general formula (I) or (II) may be produced by reacting an alkenyl or alkyl succinic acid or alkenyl or alkyl succinic anhydride with a corresponding amine.

[0029] As the amine, there may be mentioned, for example, ethylenediamine, propanediamine, butanediamine, N-methyl-1,3-propanediamine, N,N-dimethyl-1,3-propanediamine and a polyalkylenepolyamine such as diethylenetriamine, triethylenetetramine, aminoethylpiperazine or tetraethylenepentamine.

[0030] The boron-containing succinimide compound may be obtained by reacting the boron-free succinimide compound with a boron-containing compound generally at a temperature of 50 to 250°C, preferably 100 to 200°C.

[0031] As the boron-containing compound, there may be used at least one compound selected from a boron oxide, a boron halide, boric acid, boric anhydride, a borate ester, etc.

[0032] In the present invention, as the raw material (A), the above-described succinimide compounds may be used singly or in combination with two or more thereof.

[0033] As the phosphorus atom-containing compound used as the raw material (B), there may be mentioned, for example, a phosphorus sulfide, a phospho sulfurized hydrocarbon compound, a phosphate ester, a phosphite ester and a dithiophosphate ester.

[0034] The alkyl group or groups incorporated into the phosphorus atom-containing compound may be linear or branched and may contain a cyclic structure. Further, an ether bond and/or a thioether bond may be also contained in the alkyl group or groups.

[0035] As the phosphorus sulfide, there may be mentioned P_2S_3 , P_2S_5 , P_4S_7 and P_4S_{10} . Above all, P_2S_5 is preferred.

[0036] As the phospho sulfurized hydrocarbon compound, there may be used various compounds. Phospho sulfurized hydrocarbon compounds having a structure in which two alkyl groups are bonded to a phosphorus atom are preferable.

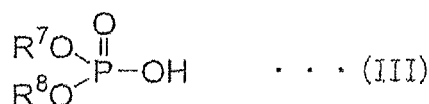
[0037] Such compounds may include reaction products of an olefin and a phosphorus sulfide.

[0038] For example, a phospho sulfurized hydrocarbon compound may be obtainable by using propylene, butene, isobutylene, a polymer or copolymer thereof, decene, cetene, octadecene, a terpene (such as pinene), vinylbornene or camphene as an olefin and P_2S_3 , P_2S_5 , P_4S_7 or P_4S_{10} as a phosphorus sulfide, and by reacting them at a temperature of about 100 to 300°C under a stream of nitrogen gas.

[0039] In this case, as raw materials, the particularly preferred olefin is α -pinene and the preferred phosphorus sulfide is P_2S_5 .

[0040] It is preferred that the phosphate ester, phosphite ester and dithiophosphate ester be each a diester.

[0041] As the preferable phosphate ester, there may be mentioned an acidic phosphate ester represented by the general formula (III):



wherein R^7 and R^8 each represent a hydrocarbonyl group having 1 to 20 carbon atoms. Specifically, R^7 and R^8 may be a linear or branched alkyl group having 1 to 20 carbon atoms, a cycloalkyl group having 3 to 20 carbon atoms, a linear or branched alkenyl group having 2 to 20 carbon atoms, an aryl group having 6 to 20 carbon atoms or an aralkyl group having 7 to 20 carbon atoms.

[0042] Examples of the alkyl group having 1 to 20 carbon atoms include a methyl group, an ethyl group, a n-propyl group, an isopropyl group, a n-butyl group, an isobutyl group, a sec-butyl group, a tert-butyl group, a pentyl group, a hexyl group, an octyl group, a 2-ethylhexyl group, a decyl group, a dodecyl group, a tetradecyl group, a hexadecyl group, an octadecyl group and an eicosyl group.

[0043] Examples of the cycloalkyl group having 3 to 20 carbon atoms include a cyclopentyl group, a cyclohexyl group, a methylcyclohexyl group and a cyclooctyl group.

[0044] Examples of the alkenyl group having 2 to 20 carbon atoms include an allyl group, a propenyl group, a butenyl group, an octenyl group, a decenyl group and an oleyl group.

[0045] Examples of the aryl group having 6 to 20 carbon atoms include a phenyl group, a tolyl group, a xylyl group and a naphthyl group. Examples of the aralkyl group having 7 to 20 carbon atoms include a benzyl group, a phenethyl group and a naphthylmethyl group.

[0046] The hydrocarbonyl group having 1 to 20 carbon atoms may be interrupted by one or more oxygen atoms and/or sulfur atoms.

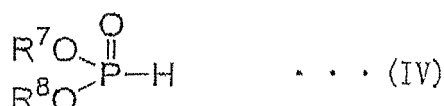
[0047] Namely, the main chain may contain at least one ether bond or thioether bond or contain both ether and thioether

bonds.

[0048] Examples of such a hydrocarbonyl group include a hexyloxymethyl group, a hexyloxyethyl group, an octyloxymethyl group, an octyloxyethyl group, a dodecyloxymethyl group, a dodecyloxyethyl group, a hexadecyloxymethyl group, a hexadecyloxyethyl group, a hexylthiomethyl group, a hexylthioethyl group, an octylthiomethyl group, an octylthioethyl group, a dodecylthiomethyl group, a dodecylthioethyl group, a hexadecylthiomethyl group and a hexadecylthioethyl group.

[0049] As the acidic phosphate ester represented by the general formula (III), there may be mentioned, for example, dihexyl phosphate, dioctyl phosphate, di(2-ethylhexyl) phosphate, didodecyl phosphate, dihexadecyl phosphate, di(hexylthioethyl) phosphate, di(octylthioethyl) phosphate, di(dodecylthioethyl) phosphate, di(hexadecylthioethyl) phosphate, dioctenyl phosphate, dioleyl phosphate, dicyclohexyl phosphate, diphenyl phosphate, ditolyl phosphate, dibenzyl phosphate and diphenethyl phosphate.

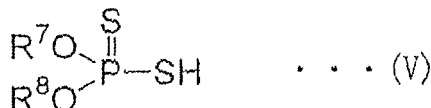
[0050] As the preferred phosphite ester, there may be mentioned an acidic phosphite ester represented by the general formula (IV):



wherein R^7 and R^8 are as defined above.

[0051] As the acidic phosphite ester represented by the general formula (IV), there may be mentioned, for example, dihexyl hydrogen phosphite, dioctyl hydrogen phosphite, di(2-ethylhexyl) hydrogen phosphite, didodecyl hydrogen phosphite, dihexadecyl hydrogen phosphite, di(hexylthioethyl) hydrogen phosphite, di(octylthioethyl) hydrogen phosphite, di(dodecylthioethyl) hydrogen phosphite, di(hexadecylthioethyl) hydrogen phosphite, dioctenyl hydrogen phosphite, dioleyl hydrogen phosphite, dicyclohexyl hydrogen phosphite, diphenyl hydrogen phosphite, ditolyl hydrogen phosphite, dibenzyl hydrogen phosphite and diphenethyl hydrogen phosphite.

[0052] As the preferred dithiophosphate ester, there may be mentioned a dithiophosphate ester represented by the general formula (V):



wherein R^7 and R^8 are as defined above.

[0053] As the dithiophosphate ester represented by the general formula (V), there may be mentioned, for example, dihexyl dithiophosphate, dioctyl dithiophosphate, di(2-ethylhexyl) dithiophosphate, didodecyl dithiophosphate, dihexadecyl dithiophosphate, di(hexylthioethyl) dithiophosphate, di(octylthioethyl) dithiophosphate, di(dodecylthioethyl) dithiophosphate, di(hexadecylthioethyl) dithiophosphate, dioctenyl dithiophosphate, dioleyl dithiophosphate, dicyclohexyl dithiophosphate, diphenyl dithiophosphate, ditolyl dithiophosphate, dibenzyl dithiophosphate and diphenethyl dithiophosphate.

[0054] In the present invention, as the raw material (B), the above-described phosphorus atom-containing compounds may be used singly or in combination with two or more thereof.

[0055] The lubricating oil additive of the present invention is a reaction product obtained by reacting the above-described raw material (A) with raw material (B) in a molar ratio of preferably 1:10 to 10:1, more preferably 1:3 to 3:1. By using the raw material (A) and raw material (B) in the above proportion range, a lubricating oil additive having the desired performance may be obtained.

[0056] The reaction may be carried out at a temperature of generally about 50 to 250°C, preferably 100 to 200°C.

[0057] In this case, an organic solvent such as a hydrocarbon compound may be used, if necessary.

[0058] The lubricating oil additive of the present invention thus obtained can afford a lubricating oil composition, such as an automatic transmission fluid or a continuously variable transmission fluid, having a particularly high wet friction material torque capacity and excellent μ -V performances.

[0059] The lubricating oil composition of the present invention comprises a mineral oil and/or a synthetic base oil, and the above-described lubricating oil additive. The content of the lubricating oil additive in the lubricating oil composition is preferably 0.01 to 50 % by mass, more preferably 0.1 to 20 % by mass, from the standpoint of capability of effectively showing the above effects.

[0060] As the base oil used in the composition of the present invention, both mineral oil and synthetic oil may be used.

[0061] As the mineral oil, any of various conventionally known mineral oils may be used and there may be mentioned, for example, a paraffinic mineral oil, an intermediate mineral oil and a naphthenic mineral oil. Specific examples of the mineral oil include solvent-refined or hydrogen-refined light neutral oil, medium neutral oil, heavy neutral oil or bright stock and the like.

[0062] As the synthetic oil, any of various conventionally known synthetic oils may be used and there may be used, for example, poly(α -olefin) (including a copolymer of α -olefin), polybutene, a polyol ester, an ester of a dibasic acid, a phosphate ester, a polyphenyl ether, an alkylbenzene, an alkylnaphthalene, a polyoxyalkylene glycol, neopentyl glycol, silicone oil, trimethylol propane, pentaerythritol and, further, a hindered ester.

[0063] These base oils may be used singly or in combination of two or more thereof. A mineral oil and a synthetic oil may be also used in combination.

[0064] It is preferred that the base oil have a kinematic viscosity at 100°C of 1 to 30 mm²/s and %C_A of 20 % or less.

[0065] When the kinematic viscosity is in the above range, it is possible to sufficiently reduce the friction in a sliding part, such as a gear bearing or a clutch, of an automatic transmission. Additionally, the low-temperature characteristics are good.

[0066] The kinematic viscosity at 100°C is more preferably 2 to 20 mm²/s, particularly preferably 3 to 10 mm²/s.

[0067] When the %C_A is 20 % or less, the low-temperature characteristics are good. The %C_A is particularly preferably 10 % or less.

[0068] Various additives other than the lubricating oil additive of the present invention may be incorporated, singly or in combination of a plurality thereof, into the lubricating oil composition of the present invention for the purpose of further improving the performance thereof. Representatives of such additives include an ashless dispersant, a metallic detergent, a friction modifier, a viscosity index improver, an extreme pressure additive, a corrosion inhibitor, an antifoaming agent and a colorant. The lubricating oil additive of the present invention would not hinder the effects of such other additives.

[0069] The lubricating oil composition of the present invention thus formulated is suited as an automatic transmission fluid or a continuously variable transmission fluid having a high wet friction material torque capacity and excellent μ -V performances and may be also used as a lubricating oil for construction or agricultural machines equipped with a transmission having a wet clutch or a wet brake, manual transmissions, motorcycle gasoline engines, diesel engines, gas engines and shock absorbers.

[0070] The present invention will be next described by Examples but is not limited thereto in any way.

[0071] In Examples, the performance of the lubricating oil composition was evaluated according to the following method. (1) Evaluation of Wet Friction Material Torque Capacity and μ -V Performances:

Using a Low Velocity Friction Apparatus, the test was carried out under the conditions shown below to evaluate the wet friction material torque capacity in terms of a friction coefficient at 250 rpm (μ_{250}). The μ -V performances were evaluated in terms of μ ratio (μ_{50}/μ_1).

The higher the μ_{250} value is, the higher the torque capacity is. A μ_{50}/μ_1 value of greater than 1 is regarded as being good. Designated as μ_1 is a friction coefficient at 1 rpm, while as μ_{50} is a friction coefficient at 50 rpm.

Test conditions:

Friction material:	commercially available cellulose wet paper material and steel plate
Test temperature:	120°C
Surface pressure:	0.98 MPa
Measurement:	1 to 300 rpm, stepwise

Example 1

(1) Preparation of Lubricating Oil Additive

[0072] In a 500 mL separable flask, 0.12 mole of borated succinimide (mono-type, alkyl group: polybutene having an average molecular weight of 960, boron content: 1.0 % by mass; unless otherwise specifically noted, similar succinimide was used in the following examples) and 0.17 mole of diolel hydrogen phosphite were charged and reacted at 150°C for 6 hours under a stream of nitrogen gas. The water by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (a) which was a reaction product of borated succinimide and diolel hydrogen phosphite.

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0073] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 87.8 % by mass of the mineral oil and 12.2 % by mass of the lubricating oil additive (a) obtained in (1) above was prepared, so

that the content of nitrogen derived from the lubricating oil additive (a) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 2

(1) Preparation of Lubricating Oil Additive

[0074] A lubricating oil additive (b) was prepared in the same manner as that described in Example 1-(1) except that di(2-ethylhexyl) hydrogen phosphite was substituted for diolel hydrogen phosphite in Example 1-(1).

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0075] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 91.7 % by mass of the mineral oil and 8.3 % by mass of the lubricating oil additive (b) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (b) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 3

(1) Preparation of Lubricating Oil Additive

[0076] A lubricating oil additive (c) was prepared in the same manner as that described in Example 1-(1) except that di(octylthioethyl) hydrogen phosphite was substituted for diolel hydrogen phosphite in Example 1-(1).

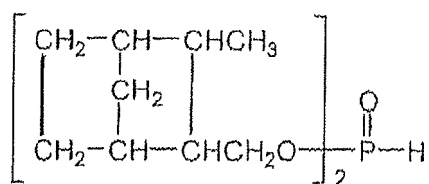
(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0077] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 90.0 % by mass of the mineral oil and 10.0 % by mass of the lubricating oil additive (c) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (c) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 4

(1) Preparation of Lubricating Oil Additive

[0078] A lubricating oil additive (d) was prepared in the same manner as that described in Example 1-(1) except that a phosphorus compound represented by the following formula



was substituted for diolel hydrogen phosphite in Example 1-(1).

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0079] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 90.6 % by mass of the mineral oil and 9.4 % by mass of the lubricating oil additive (d) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (d) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 5

(1) Preparation of Lubricating Oil Additive

5 **[0080]** In a 500 mL separable flask, 0.12 mole of borated succinimide (boron content: 0.4 % by mass) and 0.06 mole of phospho sulfurized pinene were charged and reacted at 170°C for 8 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (e) which was a reaction product of borated succinimide and phospho sulfurized pinene.

10 (2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0081] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 89.7 % by mass of the mineral oil and 10.3 % by mass of the lubricating oil additive (d) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (e) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 6

(1) Preparation of Lubricating Oil Additive

20 **[0082]** A lubricating oil additive (f) was prepared in the same manner as that described in Example 5-(1) except that di(2-ethylhexyl) dithiophosphate was substituted for phospho sulfurized pinene in Example 5-(1).

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

25 **[0083]** Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 92.1 % by mass of the mineral oil and 7.9 % by mass of the lubricating oil additive (f) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (f) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 7

(1) Preparation of Lubricating Oil Additive

35 **[0084]** In a 500 mL separable flask, 0.12 mole of borated succinimide (boron content: 1.0 % by mass) and 0.04 mole of diphosphorus pentasulfide were charged and reacted at 170°C for 8 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (g) which was a reaction product of borated succinimide and diphosphorus pentasulfide.

40 (2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0085] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 91.2 % by mass of the mineral oil and 8.8 % by mass of the lubricating oil additive (g) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (g) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 8

(1) Preparation of Lubricating Oil Additive

50 **[0086]** A lubricating oil additive (h) was prepared in the same manner as that described in Example 5-(1) except that borated succinimide (boron content: 0.6 % by mass) was substituted for borated succinimide (boron content: 0.4 % by mass) in Example 5-(1).

55 (2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0087] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 92.5 % by mass of the mineral oil and 7.5 % by mass of the lubricating oil additive (h) obtained in (1) above was prepared, so

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that the content of nitrogen derived from the lubricating oil additive (h) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 9

(1) Preparation of Lubricating Oil Additive

[0088] In a 200 mL separable flask, 50 g of commercially available borated succinimide (boron content: 2.0 % by mass) and 5.5 g of phospho sulfurized pinene were charged and reacted at 170°C for 8 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (i).

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0089] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 93.2 % by mass of the mineral oil and 6.8 % by mass of the lubricating oil additive (i) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (i) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 10

(1) Preparation of Lubricating Oil Additive

[0090] In a 300 mL four-necked flask, a solution of 0.035 mole of borated succinimide (boron content: 1.0 % by mass) in 70 mL of xylene was prepared, to which a solution of 0.023 mole of diphosphorus pentasulfide in 70 mL of xylene was added at 100°C under a stream of nitrogen gas.

[0091] The mixture was reacted at 160°C for 6 hours under reflux. Thereafter, xylene was removed by distillation, thereby obtaining a lubricating oil additive (j) which was a reaction product of borated succinimide and diphosphorus pentasulfide.

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0092] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 92.5 % by mass of the mineral oil and 7.5 % by mass of the lubricating oil additive (j) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (j) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Example 11

(1) Preparation of Lubricating Oil Additive

[0093] A lubricating oil additive (k) was prepared in the same manner as that described in Example 10-(1) except that borated succinimide (boron content: 0.6 % by mass) was substituted for borated succinimide (boron content: 1.0 % by mass) in Example 10-(1).

(2) Preparation of Lubricating Oil Composition and Evaluation of its Performance:

[0094] Using a mineral oil corresponding to 150 neutral as a base oil, a lubricating oil composition containing 92.1 % by mass of the mineral oil and 7.9 % by mass of the lubricating oil additive (k) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (k) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 1.

Table 1

	Lubricating Oil Composition			Evaluation of Performance	
	Mineral oil (% by mass)	Lubricating oil additive		Torque capacity [μ 250]	μ ratio [μ 50/ μ 1]
		Kind	(% by mass)		
Example 1	87.8	(a)	12.2	0.164	1.293

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(continued)

	Lubricating Oil Composition			Evaluation of Performance	
	Mineral oil (% by mass)	Lubricating oil additive		Torque capacity [μ 250]	μ ratio [μ 50/ μ 1]
		Kind	(% by mass)		
Example 2	91.7	(b)	8.3	0.173	1.219
Example 3	90.0	(c)	10.0	0.163	1.197
Example 4	90.6	(d)	9.4	0.166	1.197
Example 5	89.7	(e)	10.3	0.188	1.105
Example 6	92.1	(f)	7.9	0.176	1.267
Example 7	91.2	(g)	8.8	0.180	1.128
Example 8	92.5	(h)	7.5	0.190	1.108
Example 9	93.2	(i)	6.8	0.186	1.046
Example 10	92.5	(j)	7.5	0.193	1.066
Example 11	92.1	(k)	7.9	0.183	1.058

Comparative Examples 1 to 5

[0095] Using a mineral oil corresponding to 150 neutral as a base oil and an additive (1) or additives (1) and (2) shown in Table 2 as a lubricating oil additive, lubricating oil compositions having the formulations shown in Table 3 were prepared, so that the content of nitrogen derived from the additive (1) was 1,500 ppm by mass. The performance of each of the compositions was evaluated. The results are shown in Table 3.

Table 2

	Additive 1	Additive 2
Comparative Example 1	Monosuccinimide	-
Comparative Example 2	Bissuccinimide	-
Comparative Example 3	Borated succinimide (boron content: 2.0 % by mass)	-
Comparative Example 4	Borated succinimide (boron content: 1.0 % by mass)	Diolel hydrogen phosphite
Comparative Example 5	Borated succinimide (boron content: 1.0 % by mass)	Phospho sulfurized pinene

Table 3

	Lubricating Oil Composition			Evaluation of Performance	
	Mineral oil (% by mass)	Additive (1) (% by mass)	Additive (2) (% by mass)	Torque capacity [μ 250]	μ ratio [μ 50/ μ 1]
Comparative Example 1	92.0	8.0	-	0.129	0.960
Comparative Example 2	84.7	15.3	-	0.114	0.925
Comparative Example 3	93.2	6.8	-	0.155	0.966
Comparative Example 4	89.9	7.5	2.6	0.147	1.545
Comparative Example 5	91.7	7.5	0.8	0.159	1.067

[0096] As will be evident from the above results, the lubricating oil compositions of Examples 1 to 11 containing the lubricating oil additive of the present invention can maintain the transmission torque capacity at a high level and show a μ ratio (μ_{50}/μ_1) of greater than 1.

[0097] In the case of the lubricating oil compositions containing only an imide dispersant (Comparative Examples 1 to 3), on the other hand, the transmission torque capacity is insufficient and the μ ratio is less than 1.

[0098] In the case of the lubricating oil compositions which contain borated imide and a phosphorus compound rather than a reaction product of them (Comparative Examples 4 and 5), the transmission torque capacity is insufficient.

(2) Evaluation as Automatic Transmission Fluid Composition (LVFA)

[0099] Automatic transmission fluid compositions which contained lubricating oil additives (the reaction products of borated succinimide and phospho sulfurized pinene, giving the highest μ_d value in Examples 1 to 11) and which were compounded with various additives shown in Table 4 (polymethacrylate, calcium sulfonate, isostearamide and oleyl glyceride; numerals are % by mass) were prepared and evaluated for the purpose of confirming the performance as automatic transmission fluid compositions.

[0100] As other additives, the automatic transmission fluid compositions were further compounded with 1.1 % by mass of a mixture containing 30 % by mass of a phenol antioxidant, 30 % by mass of an amine antioxidant, 30 % by mass of a sulfur antioxidant, 3 % by mass of a thiadiazole copper inactivation agent and 10 % by mass of a silicone antifoaming agent.

[0101] The automatic transmission fluid compositions were each compounded with a mixture, as a base oil, containing 12.5 % by mass of a mineral oil corresponding to 60 neutral and the balance of a mineral oil corresponding to 100 neutral.

Example 12

(1) Preparation of Lubricating Oil Additive

[0102] In a 500 mL separable flask, 0.12 mole of borated succinimide (boron content: 0.4 % by mass) and 0.06 mole of phospho sulfurized pinene were charged and reacted at 170°C for 8 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (I) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

(2) Evaluation of Performance:

[0103] An automatic transmission fluid composition containing the lubricating oil additive (I) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (I) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

Example 13

(1) Preparation of Lubricating Oil Additive

[0104] In a 200 mL separable flask, 0.06 mole of borated succinimide (boron content: 0.6 % by mass) and 0.03 mole of phospho sulfurized pinene were charged and reacted at 170°C for 8 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (m) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

(2) Evaluation of Performance:

[0105] An automatic transmission fluid composition containing the lubricating oil additive (m) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (m) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

Example 14

(1) Preparation of Lubricating Oil Additive

[0106] A lubricating oil additive (n) was prepared in the same manner as that described in Example 13-(1) except that borated succinimide (boron content: 1.0 % by mass) was substituted for borated succinimide (boron content: 0.6 % by

mass) in

Example 13-(1).

5 (2) Evaluation of Performance:

[0107] An automatic transmission fluid composition containing the lubricating oil additive (n) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (n) was 1,500 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

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Example 15

(1) Preparation of Lubricating Oil Additive

15 **[0108]** In a 200 mL separable flask, 0.031 mole of borated succinimide (boron content: 3.0 % by mass) and 0.023 mole of phospho sulfurized pinene were charged and reacted at 180°C for 6 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (o) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

20 (2) Evaluation of Performance:

[0109] An automatic transmission fluid composition containing the lubricating oil additive (o) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (o) was 750 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

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Example 16

(1) Preparation of Lubricating Oil Additive

30 **[0110]** In a 200 mL separable flask, 0.033 mole of borated succinimide (boron content: 2.8 % by mass) and 0.020 mole of phospho sulfurized pinene were charged and reacted at 180°C for 6 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (p) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

35 (2) Evaluation of Performance:

[0111] An automatic transmission fluid composition containing the lubricating oil additive (p) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (p) was 750 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

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Example 17

(1) Preparation of Lubricating Oil Additive

45 **[0112]** In a 200 mL separable flask, 0.034 mole of borated succinimide (boron content: 2.0 % by mass) and 0.017 mole of phospho sulfurized pinene were charged and reacted at 180°C for 6 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (q) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

50 (2) Evaluation of Performance:

[0113] An automatic transmission fluid composition containing the lubricating oil additive (q) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (q) was 750 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

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Example 18

(1) Preparation of Lubricating Oil Additive

[0114] In a 200 mL separable flask, 0.033 mole of borated succinimide (boron content: 2.0 % by mass) and 0.020 mole of phospho sulfurized pinene were charged and reacted at 180°C for 6 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (r) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

(2) Evaluation of Performance:

[0115] An automatic transmission fluid composition containing the lubricating oil additive (r) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (r) was 750 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

Example 19

(1) Preparation of Lubricating Oil Additive

[0116] In a 200 mL separable flask, 0.034 mole of borated succinimide (boron content: 2.4 % by mass) and 0.017 mole of phospho sulfurized pinene were charged and reacted at 180°C for 6 hours under a stream of nitrogen gas. The hydrogen sulfide by-produced was removed in vacuo at 150°C, thereby obtaining a lubricating oil additive (s) which was a reaction product of borated succinimide and phosphor sulfurized pinene.

(2) Evaluation of Performance:

[0117] An automatic transmission fluid composition containing the lubricating oil additive (s) obtained in (1) above was prepared, so that the content of nitrogen derived from the lubricating oil additive (s) was 750 ppm by mass. The performance of the composition was evaluated. The results are shown in Table 4.

Comparative Example 6

[0118] An automatic transmission fluid composition containing tricresyl phosphate, as a phosphor source, and borated succinimide in lieu of the additive of Examples 12 to 19 was prepared. The performance of the composition was evaluated.

[0119] The amount of nitrogen contained in the lubricating oil composition was 800 ppm by mass. The results are shown in Table 4.

Table 4

Automatic Transmission Fluid Composition		Example								Comparative Example
		12	13	14	15	16	17	18	19	6
Base	100 Neutral mineral oil	balance	balance	balance	balance	balance	balance	balance	balance	balance
oil	60 Neutral mineral oil	12.5	12.5	12.5	12.5	12.5	12.5	12.5	12.5	12.5
Polymethacrylate		8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5	8.5
Calcium sulfonate		1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Isostearamide		0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Oleyl glyceride		0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Borated imide		-	-	-	-	-	-	-	-	1.5
Tricresyl phosphate		-	-	-	-	-	-	-	-	0.3
Additive (l)		7.9	-	-	-	-	-	-	-	-
Additive (m)		-	7.9	-	-	-	-	-	-	-
Additive (n)		-	-	7.9	-	-	-	-	-	-
Additive (o)		-	-	-	3.1	-	-	-	-	-
Additive (p)		-	-	-	-	3.8	-	-	-	-
Additive (q)		-	-	-	-	-	4.7	-	-	-
Additive (r)		-	-	-	-	-	-	3.8	-	-
Additive (s)		-	-	-	-	-	-	-	4.7	-
Other additive (*)		1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1
Torque Capacity [μ 250]		0.156	0.174	0.167	0.166	0.169	0.170	0.170	0.173	0.146
μ Ratio [μ 50/ μ 1]		1.133	1.208	1.156	1.273	1.275	1.232	1.323	1.274	1.306
(*) An antioxidant, a copper inactivation agent and an antifoaming agent were compounded in a constant amount.										

Industrial Applicability

[0120] The lubricating oil additive of the present invention can afford a lubricating oil composition, such as an automatic transmission fluid or a continuously variable transmission fluid, having a particularly high wet friction material torque capacity and excellent μ -V performances.

[0121] The lubricating oil composition of the present invention containing the above lubricating oil additive is usable as an automatic transmission fluid or a continuously variable transmission fluid and as a lubricating oil for construction or agricultural machines equipped with a transmission having a wet clutch or a wet brake, manual transmissions, motorcycle gasoline engines, diesel engines, gas engines and shock absorbers.

Claims

1. A lubricating oil additive comprising a reaction product of a succinimide compound and a phosphorus atom-containing compound.
2. A lubricating oil additive as recited in claim 1, wherein the phosphorus atom-containing compound is at least one compound selected from the group consisting of phosphorus sulfide, a phospho sulfurized hydrocarbon compound, a phosphate ester, a phosphite ester and a dithiophosphate ester.
3. A lubricating oil additive as recited in claim 2, wherein the phospho sulfurized hydrocarbon compound has a structure in which two alkyl groups are bonded to a phosphorus atom.
4. A lubricating oil additive as recited in claim 2, wherein the phosphate ester, phosphite ester and dithiophosphate ester are each a diester.
5. A lubricating oil additive as recited in claim 2, wherein the alkyl group introduced into the phosphorus atom-containing hydrocarbon compound is an alkyl group which has 2 to 25 carbon atoms and which may have an ether bond and/or a thioether bond.
6. A lubricating oil composition **characterized by** comprising a mineral oil and/or a synthetic base oil, and a lubricating oil additive according to claim 1.
7. A lubricating oil composition as recited in claim 6, wherein the composition is used for a transmission having a wet clutch or a wet brake.
8. A lubricating oil composition as recited in claim 6, wherein the composition is an automatic transmission fluid or a continuously variable transmission fluid.

INTERNATIONAL SEARCH REPORT

International application No.

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A. CLASSIFICATION OF SUBJECT MATTER Int.Cl ⁷ C10M159/12// (C10M159/12, 125:24, 133:16, 137:00, 139:00), C10N30:00, 40:04, 60:00 According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) Int.Cl ⁷ C10M159/12, 125/24, 133/16, 133/56, 137/00-137/16, 139/00, C10N30:00, 40:04, 60:00 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922-1996 Toroku Jitsuyo Shinan Koho 1994-2004 Kokai Jitsuyo Shinan Koho 1971-2004 Jitsuyo Shinan Toroku Koho 1996-2004 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
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* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "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) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "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 "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "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 "&" document member of the same patent family		
Date of the actual completion of the international search 20 December, 2004 (20.12.04)		Date of mailing of the international search report 11 January, 2005 (11.01.05)
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