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(54) **Lubricating oil additive composition having a high friction coefficient at elevated temperatures**

(57) The present invention provides a lubricating oil additive composition containing a salt of an ashless dispersant and an aromatic carboxylic acid having from about 1 to 4 carboxylic groups when employed as lubri-

cant additives provides a high friction coefficient at elevated temperatures. The lubricating oil composition containing the present additive composition is especially useful in hydraulic oils and automatic transmission oils.

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Description

[0001] The present invention relates to a novel lubricating oil additive composition. In a further aspect, the present invention relates to a novel lubricating oil additive composition having a high friction coefficient at elevated temperatures when employed in a lubricating oil composition. In still a further aspect, the present invention relates to a process for improving the friction coefficient of a lubricating oil composition at elevated temperatures.

BACKGROUND OF THE INVENTION

[0002] Modern lubricating oil compositions are formulated to exacting specifications often set by original equipment manufacturers. To meet such specifications, various additives are used, together with base oil of lubricating viscosity. Depending on the application, a typical lubricating oil composition may contain dispersants, detergents, anti-oxidants, wear inhibitors, rust inhibitors, corrosion inhibitors, foam inhibitors just to name a few. Different applications will govern the type of additives that will go into a lubricating oil composition.

[0003] Hydraulic oils and automatic transmission oils, for example, have very specific friction requirements. For instance, these lubricating oils require high friction coefficients. For example, hydraulic oil used for disc brakes must have a high friction coefficient to be effective. Further, automatic transmission oil must have enough friction for the clutch plates to transfer power. However, the friction coefficient of oil has a tendency to decline due to the temperature effects as the oil heats up during operation. It is important that the hydraulic oil or automatic transmission oil maintain its high friction coefficient at elevated temperatures, otherwise brake systems or automatic transmissions may fail.

[0004] Friction coefficient modifiers have been used in an attempt to maintain the frictional performance of hydraulic oil or automatic transmission oil. Examples of friction coefficient modifiers include higher fatty acids, higher alcohols, aliphatic amines, aliphatic amides, aliphatic esters, sulfurized oils or fats, acidic phosphate esters, acidic phosphite esters, oil soluble organic molybdenum compounds and solid lubricants. These additives have improved good service life of such oils, but none provide high friction coefficient to a lubricating oil composition at elevated temperatures.

SUMMARY OF THE INVENTION

[0005] The present invention is based on the surprising discovery that a lubricating oil additive composition containing a salt comprising an ashless dispersant and an aromatic carboxylic acid provides a high friction coefficient at elevated temperatures, particularly above about 80°C, when employed in a lubricating oil composition.

[0006] In one embodiment, the lubricating oil additive composition contains a salt comprising a polyisobutenylsuccinimide and an aromatic carboxylic acid having from about 1 to 4 carboxylic acid groups in a molar ratio ranging from about 0.9:1 to 10:1.

[0007] The polyisobutenylsuccinimide is prepared by reacting a polyalkylene polyamine and polyisobutenyl succinic anhydride under reactive conditions, wherein the polyisobutenyl group has an average molecular weight in the range of about 500 to 5,000. Suitable polyalkylene polyamines include ethylenediamine, diethylenetriamine, triethylenetetramine, and tetraethylenepentamine. The polyisobutenyl-succinimide may be further modified by post-treatment with a boric acid or cyclic carbonate.

[0008] The aromatic carboxylic acid is preferably selected from a group consisting of benzoic acid, phthalic acid, isophthalic, terephthalic acid, trimellitic acid, and pyromellitic acid and is in an amount in the range of about 0.01 to 10 wt %, based on the total weight of the lubricating oil composition.

[0009] In a further embodiment, the present invention provides for a lubricating oil composition having a major amount of a base oil of lubricating viscosity, and an effective friction coefficient improving amount of the lubricating oil additive composition of the present invention (generally from about 0.1 to 30 wt %). Other additives may be employed in the present invention. For example, the lubricating oil composition of the present invention may further comprise at least one of the following: zinc dialkyldithiophosphate (0 to 2 wt %), phenolic antioxidant (0 to 2 wt %), zinc dinonylnaphthalene sulfonate (0 to 1 wt %), calcium alkylsalicylate (0 to 5 wt %), alkylphenol calcium salt (0 to 5 wt %), polypropylene glycol (0 to 0.5 wt %), and glycerol monooleate (0 to 1 wt %).

[0010] The present invention is additionally directed to a process of improving the friction coefficient of a lubricating oil at elevated temperatures by adding to that lubricating oil an effective amount of the lubricating oil additive composition of the present invention.

[0011] Among other factors, the present invention is based on the surprising discovery that a salt of a polyisobutenylsuccinimide dispersant and an aromatic carboxylic acid when employed as lubricant additives provides a high friction coefficient at elevated temperatures. The lubricating oil composition containing the present additive composition is especially useful in hydraulic oils and automatic transmission oils.

DETAILED DESCRIPTION OF THE INVENTION

[0012] In its broadest aspect, the present invention involves a lubricating oil additive composition containing a salt of an ashless dispersant and an aromatic carboxylic acid. The ashless dispersant alone does not yield a high friction coefficient at elevated temperatures. However, the lubricating oil additive composition of the present invention surprisingly provides a high friction coefficient at elevated temperatures. The lubricating oil additive composition of the present invention provides a high friction coefficient at elevated temperatures, particularly above about 80°C, when employed in a lubricating oil composition. The lubricating oil composition containing the present lubricating oil additive composition is especially useful in hydraulic oil and automatic transmission oils.

Base Oil of Lubricating Viscosity

[0013] The base oil of lubricating viscosity used in the present invention may be mineral oils, vegetable oils or synthetic oils of suitable viscosity. The base oils may be derived from synthetic or natural sources.

[0014] Mineral oils for use as the base oil in this invention include, for example, paraffinic, naphthenic and other oils that are ordinarily used in lubricating oil compositions. The mineral oil employable for the present invention may have a viscosity of 1 to 1,000 cSt, preferably 2 to 100 cSt, more preferably 2 to 50 cSt, at 40°C and can be obtained by solvent extraction or by hydrogenation.

[0015] Examples of vegetable oils include rapeseed oil, cottonseed oil, and castor oil.

[0016] Synthetic oils include, for example, both hydrocarbon synthetic oils and synthetic esters and mixtures thereof having desired viscosity. Useful synthetic hydrocarbon oils include liquid polymers of alpha olefins having the proper viscosity. Especially useful are the hydrogenated liquid oligomers of C₆ to C₁₂ alpha olefins such as 1-decene trimer. Likewise, alkyl benzenes of proper viscosity, such as didodecyl benzene, can be used. Useful synthetic esters include the esters of monocarboxylic acids and polycarboxylic acids, as well as mono-hydroxy alkanols and polyols. Typical examples are didodecyl adipate, pentaerythritol tetracaprate, di-2-ethylhexyl adipate, dilaurylsebacate, and the like. Complex esters prepared from mixtures of mono and dicarboxylic acids and mono and dihydroxy alkanols can also be used. Blends of mineral oils with synthetic oils are also useful.

Polyisobutenylsuccinimide Dispersant

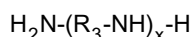
[0017] The polyisobutenylsuccinimide dispersant used in the present invention can be prepared by conventional processes. In brief, the polyisobutenyl-succinimide dispersant is derived from a polyisobutenyl succinic anhydride, wherein the polyisobutenyl group has an average molecular weight in the range of about 500 to 5,000, preferably about 700 to 3,000, more preferably about 900 to 2,500, and most preferably about 950.

[0018] The polyisobutenylsuccinimide employed in the present invention are prepared by reacting a polyisobutenyl succinic anhydride with a polyalkylene polyamine, including alkylene diamine.

[0019] Polyisobutenyl succinic anhydrides are well-known in the art and are prepared by the thermal reaction of polyisobutene and maleic anhydride as described, for example, in U.S. Pat. Nos. 3,361,673 and 3,676,089. Alternatively, polyisobutenyl succinic anhydrides can be prepared by reaction of chlorinated polyisobutene with maleic anhydride as described, for example, in U.S. Pat. No. 3,172,892. The polyisobutene employed in these reactions is commercially available and has an average molecular weight in the range of about 500 to 5,000, preferably, about 700 to 3,000, more preferably about 900 to 2,500, and most preferably about 950.

[0020] The reaction of a polyamine with an alkenyl or alkyl succinic anhydride to produce a polyamino alkenyl or alkyl succinimide is well known in the art and is described, for example, in U.S. Patent Nos. U.S. Patent No. 2,992,708; 3,018,250; 3,018,291; 3,024,237; 3,100,673; 3,172,892; 3,219,666; 3,272,746; 3,361,673; 3,381,022; 3,912,764; 4,234,435; 4,612,132; 4,747,965; 5,112,507; 5,241,003; 5,266,186; 5,286,799; 5,319,030; 5,334,321; 5,356,552; 5,716,912, the disclosures of which are all hereby incorporated by reference in their entirety for all purposes.

[0021] A particularly preferred group of polyamines for use in the present invention are polyalkylene polyamines, including alkylene diamines. Particularly suitable polyalkylene polyamines are



wherein R₃ is a straight- or branched alkylene group having from about 2 to 6 carbon atoms, preferably from about 2 to 4 carbon atoms, preferably from about 2 to 4 carbon atoms, most preferably about 2 carbon atoms, i.e. ethylene (-CH₂CH₂-); and x is an integer from about 1 to 4.

[0022] Particularly preferred polyalkylene polyamines include, but are not limited to, ethylenediamine, diethylenetriamine, triethylenetetramine, and tetraethylenepentamine. A most preferred polyalkylene polyamine is tetraethyl-

enepentamine.

[0023] The dispersancy of the polyisobutenylsuccinimide dispersant may be enhanced by further post-treatment with boric acid or a similar boron compound to form a borated polyisobutenylsuccinimide having utility within the scope of this invention. In addition to boric acid (boron acid), examples of suitable boron compounds include boron oxides, hydroxyboron halides and ester of boric acids.

[0024] The polyisobutenylsuccinimide dispersant may also be post-treated with a cyclic carbonate. The resulting modified dispersant has one or more nitrogens of the polyamino moiety substituted with a hydroxyl hydrocarbyl oxy-carbonyl, a hydroxyl poly(oxyalkylene) oxycarbonyl, a hydroxyalkylene, hydroxyalkylenepoly(oxyalkylene), or mixture thereof.

[0025] In addition to the boric acid and cyclic carbonate post-treatments, the polyisobutenylsuccinimide dispersant may be optionally post-treated with a variety of additional post-treatments designed to improve or impart different properties. Such post-treatments include those summarized in columns 27-29 of U.S. Patent No. 5,241,003. Such treatments include, treatment with:

- Inorganic phosphorous acids or anhydrides (e.g., U.S. Patent Nos. 3,403,102 and 4,648,980);
- Organic phosphorous compounds (e.g., U.S. Patent No. 3,502,677);
- Phosphorous pentasulfides;
- Boron compounds as already noted above (e.g., U.S. Patents Nos. 3,178,663 and 4,652,387);
- Carboxylic acid, polycarboxylic acids, anhydrides and/or acid halides (e.g., U.S. Patent Nos. 3,708,522 and 4,948,386);
- Epoxides polyepoxiates or thioepoxides (e.g., U.S. Patent Nos. 3,859,318 and 5,026,495);
- Aldehyde or ketone (e.g., U.S. Patent No. 3,458,530);
- Carbon disulfide (e.g., U.S. Patent No. 3,256,185);
- Glycidol (e.g., U.S. Patent No. 4,617,137);
- Urea, thiourea or guanidine (e.g., U.S. Patent Nos. 3,312,619; 3,865,813; and British Patent GB 1,065,595);
- Organic sulfonic acid (e.g., U.S. Patent No. 3,189,544 and British Patent GB 2,140,811);
- Alkenyl cyanide (e.g., U.S. Patent Nos. 3,278,550 and 3,366,569);
- Diketene (e.g., U.S. Patent No. 3,546,243);
- A diisocyanate (e.g., U.S. Patent No. 3,573,205);
- Alkane sultone (e.g., U.S. Patent No. 3,749,695);
- 1,3-Dicarbonyl Compound (e.g., U.S. Patent No. 4,579,675);
- Sulfate of alkoxyated alcohol or phenol (e.g., U.S. Patent No. 3,954,639);
- Cyclic lactone (e.g., U.S. Patent Nos. 4,617,138; 4,645,515; 4,668,246; 4,963,275; and 4,971,711);
- Cyclic carbonate or thiocarbonate linear monocarbonate or polycarbonate, or chloroformate (e.g., U.S. Patent Nos. 4,612,132; 4,647,390; 4,648,886; 4,670,170);
- Nitrogen-containing carboxylic acid (e.g., U.S. Patent 4,971,598 and British Patent GB 2,140,811);
- Hydroxyl-protected chlorodicarbonyloxy compound (e.g., U.S. Patent No. 4,614,522);
- Lactam, thiolactam, thiolactone or dithiolactone (e.g., U.S. Patent Nos. 4,614,603 and 4,666,460);
- Cyclic carbonate or thiocarbonate, linear monocarbonate or polycarbonate, or chloroformate (e.g., U.S. Patent Nos. 4,612,132; 4,647,390; 4,646,860; and 4,670,170);
- Nitrogen-containing carboxylic acid (e.g., U.S. Patent No. 4,971,598 and British Patent GB 2,440,811);
- Hydroxyl-protected chlorodicarbonyloxy compound (e.g., U.S. Patent No. 4,614,522);
- Lactam, thiolactam, thiolactone or dithiolactone (e.g., U.S. Patent Nos. 4,614,603, and 4,666,460);
- Cyclic carbamate, cyclic thiocarbamate or cyclic dithiocarbamate (e.g., U.S. Patent Nos. 4,663,062 and 4,666,459);
- Hydroxyaliphatic carboxylic acid (e.g., U.S. Patent Nos. 4,482,464; 4,521,318; 4,713,189);
- Oxidizing agent (e.g., U.S. Patent No. 4,379,064);
- Combination of phosphorus pentasulfide and a polyalkylene polyamine (e.g., U.S. Patent No. 3,185,647);
- Combination of carboxylic acid or an aldehyde or ketone and sulfur or sulfur chloride (e.g., U.S. Patent Nos. 3,390,086; 3,470,098);
- Combination of a hydrazine and carbon disulfide (e.g. U.S. Patent No. 3,519,564);
- Combination of an aldehyde and a phenol (e.g., U.S. Patent Nos. 3,649,229; 5,030,249; 5,039,307);
- Combination of an aldehyde and an O-diester of dithiophosphoric acid (e.g., U.S. Patent No. 3,865,740);
- Combination of a hydroxyaliphatic carboxylic acid and a boric acid (e.g., U.S. Patent No. 4,554,086);
- Combination of a hydroxyaliphatic carboxylic acid, then formaldehyde and a phenol (e.g., U.S. Patent No. 4,636,322);
- Combination of a hydroxyaliphatic carboxylic acid and then an aliphatic dicarboxylic acid (e.g., U.S. Patent No. 4,663,064);
- Combination of formaldehyde and a phenol and then glycolic acid (e.g., U.S. Patent No. 4,699,724);

- Combination of a hydroxyaliphatic carboxylic acid or oxalic acid and then a diisocyanate (e.g. U.S. Patent No. 4,713,191);
- Combination of inorganic acid or anhydride of phosphorus or a partial or total sulfur analog thereof and a boron compound (e.g., U.S. Patent No. 4,857,214);
- Combination of an organic diacid then an unsaturated fatty acid and then a nitrosoaromatic amine optionally followed by a boron compound and then a glycolating agent (e.g., U.S. Patent No. 4,973,412);
- Combination of an aldehyde and a triazole (e.g., U.S. Patent No. 4,963,278);
- Combination of an aldehyde and a triazole then a boron compound (e.g., U.S. Patent No. 4,981,492);
- Combination of cyclic lactone and a boron compound (e.g., U.S. Patent No. 4,963,275 and 4,971,711).

Aromatic Carboxylic Acid

[0026] The aromatic carboxylic acid has from about 1 to 4 carboxylic acid groups. Examples of the aromatic carboxylic acids include benzoic acid, phthalic acid, isophthalic acid, terephthalic acid, trimellitic acid, and pyromellitic acid. Most preferably, the aromatic carboxylic acid is isophthalic acid.

[0027] The amount of the aromatic carboxylic acid contained in the lubricating oil composition preferably is in the range of about 0.1 to 10 wt %, more preferably 0.03 to 0.1 wt %, based on the total weight of the lubricating oil composition.

[0028] Preferably, the molar ratio between the polyisobutenylsuccinimide dispersant and the aromatic carboxylic acid is from about 0.9:1 to 10:1. More preferably, the molar ratio is from about 1.5:1 to 5:1. Most preferably, the molar ratio is from about 1:1 to 2:1.

Other Additive Components

[0029] The following additive components are examples of some of the components that can be favorably employed in the present invention. These examples of additives are provided to illustrate the present invention, but they are not intended to limit it:

1. Metal detergents: sulfurized or unsulfurized alkyl or alkenyl phenates, alkyl or alkenyl aromatic sulfonates, sulfurized or unsulfurized metal salts of multi-hydroxyl alkyl or alkenyl aromatic compounds, alkyl or alkenyl hydroxyl aromatic sulfonates, sulfurized or unsulfurized alkyl or alkenyl naphthenates, metal salts of alkanolic acids, metal salts of an alkyl or alkenyl multiacid, and chemical and physical mixtures thereof.

2. Anti-oxidants: Anti-oxidants reduce the tendency of mineral oils to deteriorate in service which deterioration is evidenced by the products of oxidation such as sludge and varnish-like deposits on the metal surfaces and by an increase in viscosity. Examples of anti-oxidants useful in the present invention include, but are not limited to, phenol type (phenolic) oxidation inhibitors, such as 4,4'-methylene-bis(2,6-di-tert-butylphenol), 4,4'-bis(2,6-di-tert-butylphenol), 4,4'-bis(2-methyl-6-tert-butylphenol), 2,2'-methylene-bis(4-methyl-6-tert-butylphenol), 4,4'-butylidene-bis(3-methyl-6-tert-butylphenol), 4,4'-isopropylidene-bis(2,6-di-tert-butylphenol), 2,2'-methylene-bis(4-methyl-6-nonylphenol), 2,2'-isobutylidene-bis(4,6-dimethylphenol), 2,2'-methylene-bis(4-methyl-6-cyclohexylphenol), 2,6-di-tert-butyl-4-methylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,4-dimethyl-6-tert-butylphenol, 2,6-di-tert-butyl-4-hydroxyphenol, 2,6-di-tert-butyl-4-(N,N'-dimethylaminomethylphenol), 4,4'-thiobis(2-methyl-6-tert-butylphenol), 2,2'-thiobis(4-methyl-6-tert-butylphenol), bis(3-methyl-4-hydroxy-5-tert-butylbenzyl)-sulfide, and bis(3,5-di-tert-butyl-4-hydroxybenzyl). Diphenylamine-type oxidation inhibitors include, but are not limited to, alkylated diphenylamine, phenyl- α -naphthylamine, and alkylated- α -naphthylamine. Other types of oxidation inhibitors include metal dithiocarbamate (e.g., zinc dithiocarbamate), and methylenebis(dibutyldithiocarbamate). The anti-oxidant is generally incorporated into an engine oil in an amount of about 0 to 10 wt %, preferably 0.05 to 3.0 wt %, per total amount of the engine oil.

3. Anti-wear agents: As their name implies, these agents reduce wear of moving metallic parts. Examples of such agents include, but are not limited to, phosphates, phosphites, carbamates, esters, sulfur containing compounds, and molybdenum complexes.

4. Rust inhibitors (Anti-rust agents)

a) **Nonionic polyoxyethylene surface active agents:** polyoxyethylene lauryl ether, polyoxyethylene higher alcohol ether, polyoxyethylene nonyl phenyl ether, polyoxyethylene octyl phenyl ether, polyoxyethylene octyl stearyl ether, polyoxyethylene oleyl ether, polyoxyethylene sorbitol monostearate, polyoxyethylene sorbitol

mono-oleate, and polyethylene glycol mono-oleate.

b) Other compounds: stearic acid and other fatty acids, dicarboxylic acids, metal soaps, fatty acid amine salts, metal salts of heavy sulfonic acid, partial carboxylic acid ester of polyhydric alcohol, and phosphoric ester.

5. Demulsifiers: addition product of alkylphenol and ethylene oxide, polyoxyethylene alkyl ether, and polyoxyethylene sorbitan ester.

6. Extreme pressure agents (EP agents): zinc dialkyldithiophosphate (primary alkyl, secondary alkyl, and aryl type), sulfurized oils, diphenyl sulfide, methyl trichlorostearate, chlorinated naphthalene, fluoroalkylpolysiloxane, and lead naphthenate.

7. Friction modifiers: fatty alcohol, fatty acid, amine, borated ester, and other esters.

8. Multifunctional additives: sulfurized oxymolybdenum dithiocarbamate, sulfurized oxymolybdenum organo phosphorodithioate, oxymolybdenum monoglyceride, oxymolybdenum diethylate amide, amine-molybdenum complex compound, and sulfur-containing molybdenum complex compound.

9. Viscosity index improvers: polymethacrylate type polymers, ethylene-propylene copolymers, styrene-isoprene copolymers, hydrated styrene-isoprene copolymers, polyisobutylene, and dispersant type viscosity index improvers.

10. Pour point depressants: polymethyl methacrylate.

11. Foam inhibitors: alkyl methacrylate polymers and dimethyl silicone polymers.

Lubricating Oil Composition

[0030] The lubricating oil composition of the present invention is useful for imparting improved friction coefficient at elevated temperatures, particularly above about 80°C, when employed in a lubricating oil composition. Such a lubricating oil composition comprises a major amount of a base oil of lubricating viscosity and an effective amount of lubricating oil additive composition of the present invention. Adding an effective amount of the lubricating oil additive composition of the present invention to a lubricating oil composition provides a high friction coefficient at elevated temperatures.

[0031] In one embodiment, the lubricating oil composition contains:

- a) a major amount of base oil of lubricating viscosity; and
- b) to 30 wt % of the lubricating oil additive composition of the present invention;
- c) 0 to 2 wt % of a zinc dialkyldithiophosphate;
- d) 0 to 2 wt % of a phenolic antioxidant;
- e) 0 to 1 wt % of a zinc dinonylnaphthalene sulfonate;
- f) 0 to 5 wt % of a calcium alkylsalicylate;
- g) 0 to 5 wt % of a alkylphenol calcium salt;
- h) 0 to 0.5 wt % of a polypropylene glycol; and
- i) 0 to 1 wt % of a glycerol monooleate.

[0032] In a further embodiment, a lubricating oil composition is produced by blending a mixture of the above components. The lubricating oil composition produced by that process might have a slightly different composition than the initial mixture, because the components may interact. The components can be blended in any order and can be blended as combinations of components.

Examples of Additive Packages

[0033] Examples of the lubricating oil composition according to the invention are given below. Each of the following composition may further contain other additives such as antifoaming agent.

A. Hydraulic Oil 1		
5 10	Lubricating oil additive composition of the invention	0.01-2 wt. %
	Zinc dialkyldithiophosphate	0.4 wt. %
	Phenolic antioxidant	0.05 wt. %
	Zinc dinonylnaphthalene sulfonate	0.025 wt. %
	Sorbitan monooleate	0.025 wt. %
	Polyalkylenepolyamine-Fatty acid amine	0.02 wt. %
	Calcium alkylsalicylate	0.02 wt. %
	Phosphite antiwear agent	0.05 wt. %
Base oil		remainder

B. Hydraulic Oil 2		
15 20	Lubricating oil additive composition of the invention	0.01-2wt. %
	Zinc dialkyldithiophosphate	1.0 wt. %
	Zinc dinonylnaphthalenesulfonate	0.05 wt. %
	Alkenylsuccinic acid	0.1 wt. %
	Polyoxyalkylene alkyl ether	0.02 wt. %
	Base oil	remainder

C. Hydraulic Oil 3		
25 30 35	Lubricating oil additive composition of the invention	0.01-2 wt. %
	Zinc dialkyldithiophosphate	0.4 wt. %
	Phenolic antioxidant	0.05 wt. %
	Zinc dinonylnaphthalene sulfonate	0.025 wt. %
	Calcium alkylsalicylate	0.02 wt. %
	Alkylphenol calcium salt	0.1 wt. %
	Polypropylene glycol	0.03 wt. %
	Polymethacrylate viscosity index improver	0.4 wt. %
	Base oil	remainder

D. Hydraulic Oil 4		
40 45 50 55	Lubricating oil additive composition of the invention	0.01-2 wt. %
	Phenolic antioxidant	0.05 wt. %
	Sorbitan monooleate	0.04 wt. %
	Polypropylene glycol	0.002 wt. %
	Polyoxyalkylene alkyl ether	0.001 wt. %
	Phosphite antiwear agent	0.5 wt. %
	Amine antioxidant	0.1 wt. %
	Methylbenzotriazole	0.01 wt. %
	Dibenzylsulfide	0.05 wt. %
	Beef tallow diaminedioleate	0.015 wt. %
	Diisotridecyl adipate	0.6 wt. %
	Base oil	remainder

E. Hydraulic Oil 5		
	Lubricating oil additive composition of the invention	0.01-2wt. %
5	Phenolic antioxidant	0.05 wt. %
	Alkylphenol calcium salt	0.2 wt. %
	Amine antioxidant	0.05 wt. %
	Pinene P ₂ S ₅	0.2 wt. %
10	Alkenylsuccinic imide	0.05 wt. %
	Imidazoline derivative	0.05 wt. %
	Base oil	remainder

F. Gear Oil 1		
15	Lubricating oil additive composition of the invention	0.1-5 wt. %
	Alkylphenol calcium salt	1.5 wt. %
	Glycerol monooleate	0.3 wt. %
20	Phosphite antiwear agent	0.65 wt. %
	Amine antioxidant	0.4 wt. %
	Methylbenzotriazole	0.03 wt. %
	Alkenylsuccinic imide	4.5 wt. %
	Amine phosphate	0.5 wt. %
25	Sulfurized olefin	0.05 wt. %
	Base oil	remainder

G. Gear Oil 2		
30	Lubricating oil additive composition of the invention	0.1-5 wt. %
	Alkylphenol calcium salt	1.0 wt. %
	Glycerol monooleate	0.3 wt. %
	Phosphite antiwear agent	0.65 wt. %
35	Amine antioxidant	0.4 wt. %
	Methylbenzotriazole	0.03 wt. %
	Alkenylsuccinic imide	4.5 wt. %
	Dinonyl calcium sulfonate	0.5 wt. %
40	Amine phosphate	0.5 wt. %
	Sulfurized olefin	0.05 wt. %
	Diethanol cocoamide	0.5 wt. %
	Base oil	remainder

H. Gear Oil 3		
45	Lubricating oil additive composition of the invention	0.1-5 wt. %
	Alkylphenol calcium salt	1.0 wt. %
50	Glycerol monooleate	0.7 wt. %
	Phosphite antiwear agent	0.65 wt. %
	Amine antioxidant	0.4 wt. %
	Methylbenzotriazole	0.03 wt. %
	Alkenylsuccinic imide	4.5 wt. %
55	Dinonyl calcium sulfonate	0.5 wt. %
	Amine phosphate	0.5 wt. %
	Sulfurized olefin	0.05 wt. %

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(continued)

H. Gear Oil 3		
5	Diethanol cocoamide	0.3 wt. %
	Polymethacrylate viscosity index improver	5.0 wt. %
	Base oil	remainder

10	I. Automatic Transmission Oil 1	
	Lubricating oil additive composition of the invention	0.1-7 wt. %
	Zinc dialkyldithiophosphate	1.0 wt. %
	Phenolic antioxidant	0.2 wt. %
15	Calcium alkylsalicylate	0.7 wt. %
	Alkylphenol calcium salt	1.0 wt. %
	Calcium alkylsulfonate	1.7 wt. %
	Magnesium alkylsulfonate	2.5 wt. %
20	Amine antioxidant	0.1 wt. %
	Alkenylsuccinic imide	1.0 wt. %
	Polymethacrylate viscosity index improver	17.0 wt. %
	Base oil	remainder

25	J. Automatic Transmission Oil 2	
	Lubricating oil additive composition of the invention	0.1-7 wt. %
	Alkyldithiophosphate	0.7 wt. %
30	Polyoxyethylenealkylallyl ether	0.08 wt. %
	Methylbenzotriazole	0.03 wt. %
	Olefin sulfide	1.0 wt. %
	Potassium borate	2.0 wt. %
	Polymethacrylate viscosity index improver	17.0 wt. %
35	Base oil	remainder

40	K. Automatic Transmission Oil 3	
	Lubricating oil additive composition of the invention	0.1-7 wt. %
	Alkyldithiophosphate	0.7 wt. %
	Phenolic antioxidant	0.2 wt. %
	Alkylphenol calcium salt	0.6 wt. %
45	Calcium alkylsulfonate	0.2 wt. %
	Magnesium alkylsulfonate	0.1 wt. %
	Amine antioxidant	0.1 wt. %
	Pinene P ₂ S ₅	0.5 wt. %
	Potassium borate	2.0 wt. %
50	Polymethacrylate viscosity index improver	17.0 wt. %
	Base oil	remainder

55	L. Diesel Engine Oil 1	
	Lubricating oil additive composition of the invention	0.5-8 wt. %
	Zinc dialkyldithiophosphate	1.1 wt. %
	Calcium alkylsalicylate	2.0 wt. %

(continued)

L. Diesel Engine Oil 1		
5	Alkylphenol calcium salt	1.7 wt. %
	Calcium alkylsulfonate	1.2 wt. %
	Polyoxyethylenealkylallyl ether	0.1 wt. %
	Alkenylsuccinic imide	1.5 wt. %
	Base oil	remainder

M. Diesel Engine Oil 2		
15	Lubricating oil additive composition of the invention	0.5-8 wt. %
	Zinc dialkyldithiophosphate	1.3 wt. %
	Phenolic antioxidant	0.2 wt. %
	Alkylphenol calcium salt	3.2 wt. %
	Calcium alkylsulfonate	2.0 wt. %
20	Amine antioxidant	0.2 wt. %
	Alkenylsuccinic imide	4.0 wt. %
	Polymethacrylate viscosity index improver	3.0 wt. %
	Base oil	remainder

N. Diesel Engine Oil 3		
25	Lubricating oil additive composition of the invention	0.5-8 wt. %
	Zinc alkyldithiophosphate	1.3 wt. %
	Phenolic antioxidant	0.2 wt. %
30	Alkylphenol calcium salt	3.2 wt. %
	Calcium alkylsulfonate	2.0 wt. %
	Amine antioxidant	0.2 wt. %
	Alkenylsuccinic imide	4.0 wt. %
35	Olefin copolymer viscosity index improver	6.0 wt. %
	Base oil	remainder

O. Diesel Engine Oil 4		
40	Lubricating oil additive composition of the invention	0.5-8 wt. %
	Zinc dialkyldithiophosphate	1.6 wt. %
	Alkylphenol calcium salt	3.2 wt. %
45	Calcium alkylsulfonate	1.4 wt. %
	Magnesium alkylsulfonate	0.8 wt. %
	Alkenylsuccinic imide	9.0 wt. %
	Olefin copolymer viscosity index improver	8.0 wt. %
50	Base oil	remainder

P. Diesel Engine Oil 5		
55	Lubricating oil additive composition of the invention	0.5-8 wt. %
	Zinc dialkyldithiophosphate	1.3 wt. %
	Phenolic antioxidant	0.2 wt. %
	Alkylphenol calcium salt	3.2 wt. %

(continued)

P. Diesel Engine Oil 5	
Calcium alkylsulfonate	2.0 wt. %
Amine antioxidant	0.2 wt. %
Alkenylsuccinic imide	4.0 wt. %
Molybdenum dithiocarbamate	0.5 wt. %
Polymethacrylate viscosity index improver	3.0 wt. %
Base oil	remainder

Q. Diesel Engine Oil 6	
Lubricating oil additive composition of the invention	0.5-8 wt. %
Zinc dialkyldithiophosphate	1.3 wt. %
Phenolic antioxidant	0.2 wt. %
Alkylphenol calcium salt	3.2 wt. %
Calcium alkylsulfonate	2.0 wt. %
Amine antioxidant	0.2 wt. %
Alkenylsuccinic imide	4.0 wt. %
Molybdenum dithiophosphate	0.5 wt. %
Polymethacrylate viscosity index improver	3.0 wt. %
Base oil	remainder

EXAMPLES

[0034] The invention will be further illustrated by following examples, which set forth particularly advantageous method embodiments. While the Examples are provided to illustrate the present invention, they are not intended to limit it.

Example 1

[0035] In a flask, mineral base oil (viscosity: 20 cSt at 40°C) and polyisobutenyl-succinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, were placed and heated, and then an approximately equi-molar quantity of phthalic acid was dropwise added with stirring. The mixture was further stirred at 150°C for 4 hours to give a lubricating oil additive composition containing a salt comprising 2 mol. of polyisobutenylsuccinimide and 1 mol. of phthalic acid. The content of the salt was 4.0 wt. % in terms of the amount of phthalic acid (content of polyisobutenylsuccinimide was 50 wt. %). The mineral base oil accounts for the remaining 46 wt. %.

Example 2

[0036] The procedures of Example 1 were repeated except for using isophthalic acid in place of phthalic acid, to give a lubricating oil additive composition containing a salt comprising approximately equi-molar quantities of polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, and isophthalic acid. The content of the salt was 4.0 wt. % in terms of the amount of isophthalic acid.

Example 3

[0037] The procedures of Example 1 were repeated except for using terephthalic acid in place of phthalic acid, to give a lubricating oil additive composition containing a salt comprising approximately equi-molar quantities of polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, and terephthalic acid. The content of the salt was 4.0 wt. % in terms of the amount of terephthalic acid.

Example 4

[0038] The procedures of Example 1 were repeated except for using borated polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, in place of polyisobutenylsuccinimide, to give a lubricating oil additive composition containing a salt comprising approximately equi-molar quantities of borated polyisobutenylsuccinimide and phthalic acid. The content of the salt was 4.0 wt. % in terms of the amount of phthalic acid (the content of borated polyisobutenylsuccinimide was 60 wt. %).

Comparison Example 1

[0039] A lubricating oil additive composition containing polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, in the amount of 50 wt. % was prepared.

Comparison sample 2

[0040] A lubricating oil additive composition containing borated polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, in the amount of 60 wt. % was prepared.

Comparison Example 3

[0041] The procedures of Example 1 were repeated except for using alkenylsuccinic acid in place of phthalic acid, to prepare a lubricating oil additive composition containing a salt comprising approximately equi-molar quantities of polyisobutenylsuccinimide, derived from tetraethylenepentamine and wherein the polyisobutenyl group has an average molecular weight of about 950, and alkenylsuccinic acid. The content of the salt was 3.25 wt. % in terms of the amount of alkenylsuccinic acid.

Evaluation 1 of the Friction Coefficient of Lubricating Oil Composition**A. Preparation of Samples of Lubricating Oil Compositions****[0042]**

1) To a synthetic base oil (polyol-polyester base oil, viscosity: approx. 40 cSt at 40°C), the following additives were added to prepare a lubricating oil composition.

Additives: calcium phenate (0.2 wt. %), calcium sulfonate (0.05 wt. %), phenolic antioxidant (0.05 wt. %), amine antioxidant (0.05 wt. %), alkyldithiodiazole (0.01 wt. %), benzotriazole (0.01 wt. %), and sulfurized oil and fat (0.01 wt. %).

2) With respect to each of the above examples or comparison examples, 2 weight parts of the prepared additive composition were added to 100 weight parts of the above lubricating oil composition, to prepare samples 1-4 and 8-10. Independently, the same procedures were repeated except for adding 3, 4 or 5 weight parts of the additive composition prepared in Example 4, to prepare samples 5, 6 or 7, respectively.

B. Measurement of Friction Coefficient

[0043] The friction coefficient of each sample was measured by means of a micro-clutch tester (manufactured by Komatsu Engineering Co., Ltd) in accordance with the following method defined in KES 07.802.

[0044] In the presence of the lubricating oil composition sample, the specified disc was placed in contact with the specified plate. The pressure between the disc and the plate was set at 4 kgf/cm², and the disc was rotated at 20 r.p.m. The temperature was set to room temperature (25°C), 60°C, 80°C, 100°C, 120°C, and 140°C in order, and then the friction coefficient at each temperature was measured.

C. Results

[0045]

Table 1

Composition Sample No.	Temperature					
	25°C	60°C	80°C	100°C	120°C	140°C
1 (Ex.1)	0.1367	0.1477	0.1508	0.1522	0.1545	0.1544
2 (Ex.2)	0.1467	0.1656	0.1690	0.1683	0.1649	0.1607
3 (Ex.3)	0.1362	0.1479	0.1527	0.1530	0.1516	0.1457
4 (Ex.4)	0.1386	0.1527	0.1513	0.1467	0.1459	0.1440
5 (Ex.4)	0.1392	0.1511	0.1510	0.1486	0.1409	0.1328
6 (Ex.4)	0.1403	0.1676	0.1717	0.1686	0.1610	0.1532
7 (Ex.4)	0.1426	0.1729	0.1773	0.1782	0.1766	0.1717
8 (C.Ex.1)	0.1219	0.1484	0.1270	0.1221	0.1204	0.1200
9 (C.Ex.2)	0.1205	0.1311	0.1301	0.1289	0.1267	0.1239
10 (C.Ex.3)	0.1391	0.1482	0.1428	0.1374	0.1323	0.1272
Remark: Each sample contained a lubricating oil additive composition of the example or comparison example indicated in the parenthesis.						

[0046] The results in Table 1 indicate the following facts. The lubricating oil compositions of the invention (samples 1-7, each of which contains the lubricating oil additive composition of the invention) exhibit high friction coefficients at room temperature, and keep the high friction coefficients even at high temperatures. In contrast, the lubricating oil compositions not containing aromatic carboxylic acid (samples 8 and 9, which contain the lubricating oil additive compositions of the comparison Examples 1 and 2, respectively) exhibit low friction coefficients at room temperature, and maintain the low friction coefficients at high temperatures. Further, the lubricating oil composition containing aliphatic carboxylic acid instead of aromatic carboxylic acid (sample 10, which contains the lubricating oil additive composition of the comparison Example 3) exhibits a high friction coefficient at room temperature, but does not keep the high friction coefficient at high temperatures.

Evaluation 2 of Friction Coefficient of Lubricating Oil Composition

A. Preparation of Samples of Lubricating Oil Compositions

[0047]

1) To a mineral base oil (hydrogenated mineral base oil, viscosity: approx. 30 cSt at 40°C), the following additives were added to prepare a lubricating oil composition.

Additives: zinc dialkyldithiophosphate (0.3 wt. %), phenolic antioxidant (0.05 wt. %), zinc dinonylnaphthalene sulfonate (0.025 wt. %), calcium alkylsalicylate (0.025 wt. %), alkylphenol calcium salt (0.08 wt. %), polypropylene glycol (0.03 wt. %), and glycerol monooleate (0.15 wt. %).

2) To 100 weight parts of the above lubricating oil composition, 1 weight part of the additive composition prepared in the Example 2 was added to prepare sample 11. Independently, sample 12 was prepared from the above lubricating oil composition without adding the additive composition.

B. Measurement of the Friction Coefficient

[0048] The friction coefficient of each sample was measured by means of a micro-clutch tester (manufactured by Komatsu Engineering Co., Ltd) in accordance with the following method regulated in KES 07.802.

[0049] In the presence of the sample of lubricating oil composition, the specified disc was placed in contact with the specified plate. The pressure between the disc and the plate was set at 4 kgf/cm², and the disc was rotated at 20 r.p.m. The temperature was set at 25°C, 60°C, 80°C, 100°C, 120°C, and 140°C in order, and then the friction coefficient at each temperature was measured.

C. Results

[0050]

Table 2

Composition Sample No.	Temperature					
	25°C	60°C	80°C	100°C	120°C	140°C
11 (Ex 2)	0.1248	0.1350	0.1479	0.1503	0.1472	0.1401
12	0.1428	0.1250	0.1168	0.1064	0.0978	0.0899
Remark: The sample 11 contained the lubricating oil additive composition of the example indicated in the parenthesis.						

[0051] The results in Table 2 indicate the following facts. The lubricating oil composition of the invention (sample 11, which contains the lubricating oil additive composition of the invention) exhibits a high friction coefficient at room temperature, and keep the high friction coefficient even at high temperatures. In contrast, the lubricating oil composition not containing aromatic carboxylic acid (sample 12) exhibits a high friction coefficient at room temperature, but does not keep the high friction coefficient at high temperatures.

[0052] While the present invention has been described with reference to specific embodiments, this application is intended to cover those various changes and substitutions that may be made by those skilled in the art without departing from the spirit and scope of the appended claims.

Claims

1. A lubricating oil additive composition containing a salt comprising a polyisobutenylsuccinimide and an aromatic carboxylic acid having from about 1 to 4 carboxylic acid groups in a molar ratio ranging from about 0.9:1 to 10:1.
2. A lubricating oil additive composition according to Claim 1, wherein the molar ratio of said polyisobutenylsuccinimide and said aromatic carboxylic acid is in a range of from about 1.5:1 to 5:1.
3. A lubricating oil additive composition according to Claim 2, wherein the molar ratio of said polyisobutenylsuccinimide and said aromatic carboxylic acid is from about 1:1 to 2:1.
4. A lubricating oil additive composition according to Claim 1, wherein said polyisobutenylsuccinimide is prepared by reacting a polyalkylene polyamine and polyisobutenylsuccinic anhydride under reactive conditions.
5. A lubricating oil additive composition according to Claim 4, wherein said polyisobutenylsuccinimide is derived from a polyalkylene polyamine selected from the group consisting of ethylenediamine, diethylenetriamine, triethylenetetramine, and tetraethylenepentamine.
6. A lubricating oil additive composition according to Claim 5, wherein said polyalkylene polyamine is tetraethylenepentamine.
7. A lubricating oil additive composition according to Claim 1, wherein the polyisobutenyl group of said polyisobutenylsuccinimide has an average molecular weight of about 500 to 5,000.
8. A lubricating oil additive composition according to Claim 7, wherein said polyisobutenyl group has an average molecular weight of about 700 to 3,000.
9. A lubricating oil additive composition according to Claim 8, wherein said polyisobutenyl group has an average molecular weight of about 900 to 2,500.
10. A lubricating oil additive composition according to Claim 9, wherein said polyisobutenyl group has an average molecular weight of about 950.
11. A lubricating oil additive composition according to Claim 4, wherein said polyisobutenylsuccinimide is further treated

with a boron compound selected from the group consisting of boron oxide, boron halide, boric acid, and esters of boric acid, under reactive conditions.

5 12. A lubricating oil additive composition according to Claim 4, wherein said polyisobutenylsuccinimide is further treated with a cyclic carbonate.

10 13. A lubricating oil additive composition according to Claim 1, wherein said aromatic carboxylic acid is selected from the group consisting of benzoic acid, phthalic acid, isophthalic, terephthalic acid, trimellitic acid, and pyromellitic acid.

14. A lubricating oil additive composition according to Claim 13, wherein said aromatic carboxylic acid is an isophthalic acid.

15 15. A lubricating oil additive composition according to Claim 14, wherein said aromatic carboxylic acid is in an amount in the range of about 0.01 to 10 wt %.

16. A lubricating oil additive composition according to Claim 15, wherein said aromatic carboxylic acid is in an amount in the range of about 0.03 to 0.1 wt%.

20 17. A lubricating oil additive composition according to Claim 1, wherein said salt has a high friction coefficient at an elevated temperature above about 80°C.

18. A lubricating oil composition comprising:

25 a) a major amount of a base oil of lubricating viscosity; and

b) an effective friction coefficient improving amount of a salt comprising a polyisobutenylsuccinimide and an aromatic carboxylic acid having from about 1 to 4 carboxylic acid groups in a molar ratio ranging from about 0.9:1 to 10:1.

30 19. A lubricating oil composition according to Claim 18, wherein the molar ratio of said polyisobutenylsuccinimide and said aromatic carboxylic acid is in a range of from about 1.5:1 to 5:1.

35 20. A lubricating oil composition according to Claim 19, wherein the molar ratio of said polyisobutenylsuccinimide and said aromatic carboxylic acid is from about 1:1 to 2:1.

21. A lubricating oil composition according to Claim 18, wherein said lubricating oil composition comprises from about 0.1 to 30 wt % of component b).

40 22. A lubricating oil composition according to Claim 18, wherein said polyisobutenylsuccinimide is prepared by reacting a polyalkylene polyamine and polyisobutenylsuccinic anhydride under reactive conditions.

45 23. A lubricating oil composition according to Claim 22, wherein said polyisobutenylsuccinimide is derived from a polyalkylene polyamine selected from the group consisting of ethylenediamine, diethylenetriamine, triethylenetetramine, and tetraethylene pentamine.

24. A lubricating oil composition according to Claim 23, wherein said polyalkylene polyamine is tetrathylenepentamine.

50 25. A lubricating oil composition according to Claim 18, wherein the polyisobutenyl group of said polyisobutenylsuccinimide has an average molecular weight of about 500 to 5,000.

26. A lubricating oil composition according to Claim 25, wherein said polyisobutenyl group has an average molecular weight of about 700 to 3,000.

55 27. A lubricating oil composition according to Claim 26, wherein said polyisobutenyl group has an average molecular weight of about 900 to 3,500.

28. A lubricating oil composition according to Claim 27, wherein said polyisobutenyl group has an average molecular

weight of about 950.

29. A lubricating oil composition according to Claim 22, wherein said polyisobutenylsuccinimide is further treated with a boron compound selected from a group consisting of boron oxide, boron halide, boric acid, and esters of boric acid, under reactive conditions.

30. A lubricating oil composition according to Claim 22, wherein said polyisobutenylsuccinimide is further treated with a cyclic carbonate.

31. A lubricating oil composition according to Claim 18, wherein said aromatic carboxylic acid is selected from a group consisting of benzoic acid, phthalic acid, isophthalic, terephthalic acid, trimellitic acid, and pyromellitic acid.

32. A lubricating oil composition according to Claim 31, wherein said aromatic carboxylic acid is an isophthalic acid.

33. A process for improving the friction coefficient of a lubricating oil at elevated temperatures, said process comprising adding to said lubricating oil an effective amount of said lubricating oil additive composition according to Claim 1.

34. A lubricating oil composition comprising:

- a) a major amount of a base oil of lubricating viscosity
- b) 0.1 to 30 wt % of the lubricating oil additive composition of Claim 1;
- c) 0 to 2 wt % of a zinc dialkyldithiophosphate;
- d) 0 to 2 wt % of a phenolic antioxidant;
- e) 0 to 1 wt % of a zinc dinonylnaphthalene sulfonate;
- f) 0 to 5 wt % of a calcium alkylsalicylate;
- g) 0 to 5 wt % of a alkylphenol calcium salt;
- h) 0 to 0.5 wt % of a polypropylene glycol; and
- i) 0 to 1 wt % of a glycerol monooleate.

35. A process of producing a lubricating oil composition comprising blending the following components together:

- a) a major amount of base oil of lubricating viscosity
- b) 0.1 to 30 wt % of the lubricating oil additive composition of Claim 1;
- c) 0 to 2 wt % of a zinc dialkyldithiophosphate;
- d) 0 to 2 wt % of a phenolic antioxidant;
- e) 0 to 1 wt % of a zinc dinonylnaphthalene sulfonate;
- f) 0 to 5 wt % of a calcium alkylsalicylate;
- g) 0 to 5 wt % of a alkylphenol calcium salt;
- h) 0 to 0.5 wt % of a polypropylene glycol; and
- i) 0 to 1 wt % of a glycerol monooleate.

36. A lubricating oil composition produced by the process according to Claim 35.



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EUROPEAN SEARCH REPORT

Application Number
EP 99 30 6421

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Place of search THE HAGUE		Date of completion of the search 14 January 2000	Examiner Rotsaert, L
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EPO FORM 1503 03/82 (P04C01)



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EUROPEAN SEARCH REPORT

Application Number
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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			

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