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3,785,980

REACTION PRODUCT OF AN AMINE AND A HYDROPEROXIDIZED ETHYLENE/PROPYLENE COPOLYMER

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15 Claims

ABSTRACT OF THE DISCLOSURE

A novel polymeric lubricating oil additive is disclosed which is the reaction product of a primary or secondary amine and a hydroperoxidized atactic ethylene/propylene copolymer.

BACKGROUND OF THE INVENTION

Field of the invention

This invention relates to additives for lubricating oils used in internal combustion engines.

During ordinary operation of internal combustion engines using hydrocarbon fuels and lubricants, a number of undesirable by-products are formed in the engine, including sludge and varnish. The varnish is a lacquer-like oil-insoluble deposit which usually forms on the surfaces of the engine pistons. Sludge is a mixture of oil, carbon, water, metals, etc., whose consistency has been variously described as mud-like or mayonnaisey. Sludge forms in deposits on the pistons and/or in the crankcase, but unlike varnish, has no adhesive properties and is more readily removed than varnish. Both sludge and varnish are believed to be formed, in part, by polymerized oxidation products of the lubricating oil and fuel.

Oxidation products which form in the lubricating oil may also cause corrosion of metal parts, particularly bearings. Other by-products derived from the fuel and lubricant may form deposits on piston heads, in PCV valves, on piston rings, and in piston grooves.

For some years it has been common practice to incorporate into fuels and lubricating oils additives designed to inhibit corrosion or the formation of sludge, varnish, and other deposits or to remove such deposits as they are formed and hold them in suspension in the lubricant until such time as the contaminated lubricating oil is drained from the crankcase.

Additives have also been used to enhance the lubricating property of the lubricating oils. Additives such as viscosity index improvers and pour point depressants are in common use, particularly in the multigrade lubricating oils which must render effective lubrication over a wide range of operating temperatures and conditions.

Description of the prior art

Ethylene/propylene copolymers and their preparation are widely described in the art. Typical descriptions will be found in U.S. Pats. 2,692,257; 2,791,576; 3,153,023; and 3,300,459. Oxidation of hydrocarbon polymers is disclosed in U.S. Pats. 2,811,514; 3,110,708; and 3,232,917. Hydroperoxidation of ethylene/propylene copolymers followed by subsequent reduction of the hydroperoxide sites to hydroxyl groups is disclosed in U.S. Pats. 3,388,067, 3,404,091, and 3,404,092. Reaction of hydrocarbon or oxygenated hydrocarbon polymers with nitrogen-containing compounds is disclosed in U.S. Pats. 3,293,326; 3,350,381; and 3,481,908. U.S. Pat. 3,076,791 describes a lubricating oil additive produced by shearing an ethylene/propylene copolymer while simultaneously reacting it with an amine, in what is disclosed to be a free-radical reaction.

SUMMARY OF THE INVENTION

The composition of this invention is a novel polymer derivative which finds use as lubricating oil additive, particularly for the reduction of varnish. It is composed of carbon, hydrogen, oxygen, and nitrogen and has a thickening power in the range of 50–150 SUS, an equivalent weight based on nitrogen of 500–15,000, and an infrared spectrum containing distinguishing absorbance peaks at frequencies of 1,590–1,610 cm^{-1} , 1,660–1,675 cm^{-1} , and 1,710–1,725 cm^{-1} . This composition is the reaction product of a primary or secondary amine and a hydroperoxidized atactic ethylene/propylene copolymer. This hydroperoxidized copolymer contains a mole ratio of 50–65 parts of ethylene to 50–35 parts of propylene. It has a thickening power of 50–150 SUS, and an infrared spectrum containing distinguishing absorbance peaks at frequencies of 1,370–1,380 cm^{-1} , 1,460–1,470 cm^{-1} , 1,700–1,750 cm^{-1} , and 2,800–3,000 cm^{-1} .

DETAILED DESCRIPTION OF THE INVENTION

The compositions of this invention

Properties.—The composition of this invention is a lubricating oil additive which has been found to impart to the lubricating oil a number of desirable properties, including excellent antivarnish characteristics. In a test (described below) designed to determine the amount of varnish deposited on an 8-cylinder Ford engine, which has been found to correlate well with the well-known Sequence VB test (see R. W. Jack, Lubrication, 25, 3, 32–35 (1970)), lubricating oils containing 1.1 weight percent of the composition of this invention gave ratings higher than the same compounded oils containing 100 percent greater quantity of commercial antivarnish additive.

The polymeric compositions of this invention are extremely shear stable. In the ASTM D-2603 Sonic Shear Stability test,¹ compositions of this invention having thickening power in the range of 50–60 showed less than 1 percent viscosity loss at 210° F.

Each composition of this invention is a mixture of nitrogenated, oxygenated, derivatives of a hydrocarbon polymer. It contains only carbon, hydrogen, oxygen, and nitrogen, and is produced by reacting a primary or secondary amine with a hydroperoxidized atactic ethylene/propylene copolymer. The compositions of this invention thus consist of mixtures of numerous reaction products of the amine and the oxygenated polymer. Because of the variety of species to be found within each mixture and the difficulty of determining each individual species, the compositions are most appropriately defined in terms of overall physical properties.

The compositions of this invention have a thickening power in the range of 50–150 SUS. "Thickening power" as used herein is defined as the viscosity at 210° F. of a neutral oil having a viscosity at 100° F. of 126 SUS and at 210° F. of 41.3 SUS, containing 2.8 weight percent of the dry polymeric composition. Thickening power is thus measured as the actual viscosity of the oil due to incorporation of the polymer. Thickening power is directly related to the molecular weight of the polymer, but is used instead of molecular weight because of ease of measurement and greater practical significance of the data. While the thickening power of the compositions of this invention may be broadly defined as being in the range of 50–150 SUS, thickening power is more usually in the range

¹ Test conditions: 20 cc. sample of polymer-containing oil; 0.4–0.5 amp output current, 20 minute test run, and reference to sample of 130 neutral oil containing 10.25% of "Paratone N," a commercial polyisobutene, having a 29% viscosity loss at 210° F.

of 50–100 SUS and, preferably, in the range of 50–85 SUS.

The compositions of this invention have an equivalent weight based on nitrogen of 500–15,000, usually 1,000–7,000, and, preferably, 1,200–5,000. "Equivalent weight based on nitrogen" as used herein means the weight of polymer present for each gram-atom of nitrogen. Equivalent weight may thus be readily determined by dividing the weight of the composition by the number of gram-atoms of amine nitrogen present.

The compositions of this invention also show a characteristic infrared spectrum. This spectrum contains distinguishing absorbance peaks at frequencies of 1,590–1,610 cm^{-1} , 1,650–1,675 cm^{-1} , and 1,710–1,725 cm^{-1} . The infrared spectrum of the compositions of this invention may also contain additional absorbance peaks, notably those peaks characteristic of the atactic ethylene/propylene copolymer from which the compositions are derived.

Reactants.—The compositions of this invention are the reaction products of primary or secondary amines and hydroperoxidized atactic ethylene/propylene copolymers.

The amines useful as reaction products may be any monoamines or polyamines containing primary or secondary amino groups, or combinations thereof. Usually, the amines will be hydrocarbyl or oxygenated hydrocarbyl mono- and polyamines having 1–50 carbon atoms, 1–10 nitrogen atoms, and 0–10 oxygen atoms; these compounds will contain only carbon, hydrogen, nitrogen, and oxygen atoms. The number of carbon atoms per molecule will generally be in the range of 2–30, preferably 2–10; the number of nitrogen atoms per molecule will generally be in the range of 1–8, preferably 2–6; and the number of oxygen atoms per molecule will be in the range of 0–6, preferably 0–4. Preferred are the alkylene polyamines, especially unsubstituted alkylene polyamines containing 2–6 nitrogen atoms. Typical amines which are suitable for use as reactants include methylamine, ethylamine, n-propylamine, isopropylamine, n-butylamine, n-hexylamine, n-decylamine, n-eicosylamine, n-tetracosylamine, tert-butylamine, dimethylamine, methylethylamine, N-n-octyl-n-hexylamine, ethylene diamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine, pentaethylene hexamine, trimethylene diamine, tetramethylene diamine, hexamethylene diamine, N-n-octyl ethylene diamine, ethanolamine, n-propanolamine, tris(hydroxymethyl)methylamine, etc. Where the polymer contains a plurality of active oxygen-containing functional groups, more than one amine molecule may become attached to the polymeric chain.

The copolymer reactant is a hydroperoxidized atactic ethylene/propylene copolymer. The hydrocarbon portion of the polymer contains 45–65, usually 50–65, mole percent of ethylene and 55–35, usually 50–35, mole percent of propylene. It has a substantially regular backbone; i.e., one free of any long units of either polypropylene or polyethylene. In most cases, there are essentially no dipropylene units and no polyethylene segments of greater than three ethylene units. The polymers are atactic in that they are free of any substantial detectable crystallinity.

The copolymer reactant has a molecular weight of approximately the same order of magnitude as the composition of this invention. Molecular weight is not determined directly, however, but rather, as noted above, is a function of thickening power of the polymer. The thickening power of the polymeric reactant is in the same range as that of the nitrogen-containing product composition; i.e., 50–150 SUS, usually 50–100 SUS, and preferably 50–85 SUS. The polymeric reactant has an infrared spectrum having distinguishing absorbance peaks at frequencies of 1,370–1,380 cm^{-1} , 1,460–1,470 cm^{-1} , 1,700–1,750 cm^{-1} , and 2,800–3,000 cm^{-1} . The peak height of the 1,700–1,750 cm^{-1} peak will generally be lower than the height of the other two peaks. Other peaks

have been observed at 715–725 cm^{-1} , and 1,150–1,160 cm^{-1} .

The copolymer reactant is prepared by hydroperoxidation of an ethylene/propylene copolymer in the presence of oxygen and a free-radical initiator. Typical free-radical initiators which may be employed include ozone, cumene hydroperoxide, dicumyl peroxide, benzoyl peroxide, azobis(isobutyronitrile), sodium persulfate, diethyl peroxydicarbonate, p-methane hydroperoxide, tert-butyl hydroperoxide, nitric oxide (N_2O_4) and the like.

The hydroperoxidation is carried out in a suitable solvent such as benzene, chlorobenzene, tert-butyl benzene, and other similar materials which are relatively inert to free radical catalysis under hydroperoxidation conditions.

The oxygen source may be the peroxide free-radical initiator itself. Preferentially, however, means are provided for supplying air, oxygen, or another oxygen-containing gas as an external source of oxygen to the hydroperoxidation reaction chamber. The gas may be supplied to the chamber at any flow rate. Usually, the air or oxygen is provided at a rate sufficient to exchange all of the oxygen in the hydroperoxidation reaction chamber every few seconds. Means are provided for maintaining the chamber at an elevated temperature usually in the range of about 50–200° C., usually about 60–150° C. The actual temperature preferred will depend on the half-life of the free-radical initiator. Means are also provided for agitating the mixture of ethylene/propylene copolymer, free-radical initiator, and solvent in order to provide optimum mixing of the components.

The ethylene/propylene copolymer which is hydroperoxidized is generally an atactic ethylene/propylene copolymer containing 45–65, usually 50–65, mole percent ethylene and 55–35, usually 50–35, mole percent propylene. It will have an average molecular weight (measured by gel permeation chromatography) of at least 130,000, and usually of 180,000–500,000 or greater, preferably in the range of 220,000–300,000. It will have a thickening power of at least 175, and usually 200–800. The polymer will preferably be free of other monomers such as dienes, e.g., cyclopentadiene, butadiene, vinyl norbornene, etc., and monoenes such as styrene, α -methyl styrene, etc. Usually less than about 2 weight percent of the polymer will be present as an olefin other than propylene or ethylene, and preferably less than about 1 weight percent of such olefins will be present.

The hydroperoxidized polymer and the amine are reacted in the presence of a relatively inert solvent. Typical solvents include the hydrocarbon oil into which the product is designed to be incorporated. Alternatively, another solvent miscible in the lubricating oil and liquid at the reaction temperature may serve as the reaction medium. This would include aromatics such as mixed xylenes, as well as aliphatic oils and halogenated materials such as o-dichlorobenzene. The amine/polymer mixture will be reacted at a temperature of 100°–200° C., usually 140°–190° C., for a period of from 1–24, preferably 6–18, hours.

The lubricating oil can be any fluid of low dielectric constant, which is inert under the reaction conditions (particularly nonsaponifiable) and of lubricating viscosity. Fluids of lubricating viscosity generally have viscosities of from 35 to 50,000 SUS at 100° F. (V_{100}). The fluid medium or oil may be derived from either natural or synthetic sources. Included among the natural hydrocarbonaceous oils are paraffin base, naphthenic base and mixed base oils. Synthetic oils include polymers of various olefins, generally of from 2 to 6 carbon atoms, alkylated aromatic hydrocarbons, etc. Nonhydrocarbon oils include polyalkylene oxide, e.g., polyethylene oxide, aromatic ethers, silicones, etc. The preferred media are the hydrocarbonaceous media, both natural and synthetic, particularly those intended for use as a crankcase lubricant, the lubricating fluid should meet the viscosity stand-

ards of the Society of Automotive Engineers Recommended Practice, SAE J300a.

The additives of this invention will be present in the oil in a finished lubricant as 0.5–5.0 weight percent (on a dry polymer basis), preferably 1.0–2.5 weight percent. They may also be concentrated in oil up to 50, preferably up to 30, weight percent for convenience in shipping and storage, with dilution to the lower concentrations prior to use. Other additives, such as detergents, rust inhibitors, antioxidants, etc., may also be present in the finished lubricant. These other additives will be present in conventional concentrations.

The following two examples will illustrate typical compositions of this invention and their preparation.

EXAMPLE 1.—HYDROPEROXIDATION

Seventy grams of a commercial ethylene/propylene copolymer were dissolved in 800 cc. of benzene at reflux by rapid stirring in a 2-liter, 3-neck round-bottom flask. (The copolymer was manufactured by the Uniroyal Company and sold under the trade name "Royalene 1220"; it had an ethylene/propylene weight ratio of 54:46, a thickening power of approximately 300–400 SUS, and an infrared spectrum with major peaks at frequencies of approximately 1,375 cm^{-1} and 1,460 cm^{-1} .) A slurry of 3 grams of azobis(isobutyronitrile) in 100 cc. of benzene was added to the solution. The mixture was stirred and heated at 70° C. for 18 hours, using an air sparge of 100 ml./min. The reaction product was then filtered through Celite 545. The filtrate was determined to be a 7.8 percent polymer concentrate. The thickening power of the hydroperoxidized polymer was 75 SUS. Such a polymer shows a characteristic infrared spectrum with distinguishing peaks at frequencies of approximately 1,375 cm^{-1} , 1,460 cm^{-1} , 1,725 cm^{-1} , and 2,900 cm^{-1} .

EXAMPLE 2.—NITROGENATION

Fifty-one grams of the filtrate from Example 1 containing 4.0 grams of dry polymer were charged to a 300-ml. round-bottom flask containing 100 ml. of an aromatic solvent. This solvent was a mixture of aromatic hydrocarbons having an ASTM D-86 distillation range of 310–350° F. and an ASTM D-611 mixed aniline point of 53–60° F. The mixture was stirred with a magnetic stirrer under vacuum to strip benzene and thereafter 2 ml. of diethylene triamine were added dropwise. The mixture was maintained at 155–165° C. for 18 hours and then filtered through Celite 545. A hydrocarbon oil diluent was added and the product topped to a bottoms temperature of 190–105° C. and 0.7 ml. Hg. Unreacted amine was recovered by distillation. The mixture was then allowed to cool to 150° C. and again filtered to give a 9.3 percent polymer concentrate having a thickening power of 57.6 SUS and an equivalent weight based on nitrogen of 3,670. The infrared spectrum of this type of polymer contain distinguishing peaks at frequencies of approximately 1,375 cm^{-1} , 1,465 cm^{-1} , 1,600 cm^{-1} , 1,670 cm^{-1} , and 1,720 cm^{-1} .

Compositions of this invention produced in the manner described in Examples 1 and 2 above were evaluated for their antivarnish capabilities in a laboratory test designed to indicate their performance as lubricating oil additives in both the Ford 8 Varnish Test described below and the Sequence VB Test. In this laboratory test an oil containing the experimental antivarnish additive is rated on its ability to control the deposition on an aluminum plate of varnish resulting from the generation of varnish precursors by the reaction of nitric oxide with dicyclopentadiene in an oxygen atmosphere. The test is conducted by reacting 20 grams of the test sample, 180 grams of a paraffinic 200 neutral oil, and 8 grams of dicyclopentadiene with 2200 ml. of nitric oxide at ambient temperature in an oxygen atmosphere. After all the nitric oxide has reacted, the reaction mixture is passed dropwise at the rate of 1 ml. per minute for 45 minutes between aluminum plates heated to 135° C. The plates, after cool-

ing to ambient temperature, are washed free of oil with hexane, air-dried, and rated according to the appearance of the deposits. This rating is the reflectometer reading of light reflected from the coated plate, based on a scale of 0–100, with 0 being the reflectometer reading of dull black paper, and 100 being the reading of a polished aluminum plate; the higher the number, the better the performance. From the previously prepared calibration curve, the rating of the test oil can be directly converted to an equivalent Sequence VB or Ford piston varnish rating.

A sample of test lubricant was prepared using a 126 neutral oil into which was incorporated 0.88 weight percent of the nitrogen-containing polymeric composition (on a dry polymer basis) having an equivalent weight based on nitrogen of 1,210, 18 mM/kg. of zinc dialkyl dithiophosphate, and zero, 20, or 40 mM/kg. (as indicated) of an overbased calcium sulfonate with 11.4 percent calcium and a 9.3 base ratio. Reflectometer ratings are shown below in Table I. For comparison purposes are shown the results of the same test on samples of the 126 neutral oil containing 0.88 weight percent (dry polymer basis) of different commercial polymeric nitrogenous methacrylate varnish inhibitors, "Methacrylate A" and "Methacrylate B."

TABLE I

Sample	Thickening power, SUS	Reflector rating, mM/kg. sulfonate		
		0	20	40
Additive of this invention.....	53	43	51	60
Methacrylate A.....	63	21	35	43
Methacrylate B.....	55	23	52	56

It is apparent from these data that the compositions of this invention provide a significant degree of antivarnish character to the test oils.

The high degree of antivarnish character of the compositions of this invention is further evidenced by their performance in the Ford 8 Engine Varnish Test. The Ford 8 test uses a 302 in.³ displacement Ford V-8 engine having a 4.00 in. bore and a 3.00 in. stroke. Fuel and air feed rates are measured, and the carburetor is manually adjusted to maintain an air/fuel ratio of 15.0±0.5. The engine is run on a three-cycle test series. The conditions of each cycle are described in Table II below:

TABLE II

Cycle	Throttle	R.P.M.	Jacket temp., ° F.	Oil temp., ° F.	Cycle duration, min.
1.....	Idle.....	750	115	120	45
2.....	Wide open.....	2,500	125	175	135
3.....	do.....	2,500	170	205	60

Each series requires four hours; ordinarily four series are run consecutively without cooling the engine for a 16-hour total test run. At the end of any 4-hour or 16-hour interval, the engine may be disassembled and the piston skirts rated for varnish using the CRC Rating Manual No. 1. If desired, more severe tests can be obtained by repeating the 16-hour sequences one or more additional times. In the examples reported in Table III below, each test was run for three to eight consecutive 16-hour sequences.

Table III below illustrates the results of five different Ford 8 Engine Varnish Tests using a typical additive of this invention both with and without a commercial polyisobutenyl succinimide dispersant. The antivarnish performance of this additive is compared directly to that of a commercial polymeric nitrogenous methacrylate varnish inhibitor, "Methacrylate C," and to the succinimide alone. The base lubricant into which the antivarnish additives and dispersant were incorporated was (except for the runs using the succinimide without the nitrogenous polymeric additives) a blend of three parts of a paraffinic 200 neutral oil having a 745–960° F. boiling range and 1 part of a paraffinic 100 neutral oil hav-

ing a 670–920° F. boiling range; the blend had a viscosity index of approximately 97. This hydrocarbon oil blend contained 30 mM/kg. of an overbased calcium sulfonate, 30 mM/kg. of a carbonated sulfurized calcium alkyl phenate and 15 mM/kg. of a mixture of zinc dialkyl- and dialkaryldithiophosphate.

5. The composition of claim 4 having an equivalent weight based on nitrogen of 1,200–5,000.

6. The composition of claim 1, wherein said amine is a hydrocarbyl or oxygenated hydrocarbyl monoamine or polyamine containing only hydrogen and 1–50 carbon atoms, 1–10 nitrogen atoms and 0–10 oxygen atoms.

TABLE III

Sample	Weight percent additive	Piston varnish rating ^a						
		32 hrs.	48 hrs.	64 hrs.	80 hrs.	96 hrs.	112 hrs.	128 hrs.
Additive of this invention	2.15	9.7	9.4		8.6		8.3	7.9
Above additive plus 3 percent succinimide	1.1	9.8		9.6		9.4		8.7
Methacrylate C	2.15	8.5	7.9					
Methacrylate C plus 3 percent succinimide	2.15	9.7	9.3	9.0	8.7	8.4	7.8	
Succinimide ^b (3 percent) ^c	None	^d 9.2		^d 7.9				

^a Scale: 0–10; 10 represents a clean (i.e., varnish-free) piston.

^b The base oil for this composition was a 50:50 mixture of the 200 neutral oil mentioned above and a 350 neutral oil having a distillation range of 760–1,040° F.; other additives were 50 mM/kg. of the aforementioned overbased calcium sulfonate and 15 mM/kg. of the aforementioned zinc dialkyl dithiophosphate.

^c The succinimide was present as 3 percent concentration in the base oil mixture (see note b).

^d Average of two runs.

It is apparent from these data that in the absence of dispersant, the compositions of this invention have a significantly higher degree of antivarnish character than do compositions containing conventional nitrogenous antivarnish additives. When a commercial dispersant is present in the lubricant, the additives of this invention permit the lubricant to maintain its superior antivarnish character far longer than do the commercial nitrogenous additives. This striking superiority in the presence of dispersant occurs even though the antivarnish additive of this invention is used at one-half the concentration of the commercial additive.

The above examples and data are intended to be illustrative only. It will be apparent to those skilled in the art that there are many embodiments of the compositions described above which are within the scope and spirit of this invention.

I claim:

1. A composition of matter useful as a lubricating oil additive which comprises a polymer containing only carbon, hydrogen, oxygen, and nitrogen, and having (a) a thickening power in the range of 50–150 SUS, (b) an equivalent weight based on nitrogen of 500–15,000, and (c) an infrared spectrum containing distinguishing absorbance peaks at frequencies of 1,590–1,610 cm.⁻¹, 1,660–1,675 cm.⁻¹, and 1,710–1,725 cm.⁻¹; and which is the reaction product of an amine containing primary or secondary amino groups or combinations thereof and hydroperoxidized atactic ethylene/propylene copolymer reacted at a temperature of about 100–200° C.; said copolymer, (a) consisting of 45–65 mole percent of ethylene and 55–35 mole percent of propylene, having (b) a thickening power of 50–150 SUS, and (c) an infrared spectrum containing distinguishing absorbance peaks at frequencies of 1,370–1,380 cm.⁻¹, 1,460–1,470 cm.⁻¹, and 1,700–1,750 cm.⁻¹.

2. The composition of claim 1 having a thickening power in the range of 50–100 SUS.

3. The composition of claim 2 having a thickening power in the range of 50–85 SUS.

4. The composition of claim 1 having an equivalent weight based on nitrogen of 1,000–7,000.

7. The composition of claim 6, wherein said amine is a monoamine or polyamine containing only hydrogen and 2–30 carbon atoms, 1–8 nitrogen atoms, and 0–6 oxygen atoms.

8. The composition of claim 1, wherein said amine is an alkylene polyamine containing 2–6 nitrogen atoms.

9. The composition of claim 1, wherein said ethylene/propylene copolymer has an average molecular weight of at least 130,000 and a thickening power of at least 175.

10. The composition of claim 9, wherein said ethylene/propylene copolymer has an average molecular weight of 180,000–500,000 and a thickening power of 200–800.

11. A lubricant composition comprising a major portion of a lubricating oil and as an additive an amount of the composition of claim 1 sufficient to provide antivarnish character to said lubricant composition.

12. The lubricant composition of claim 11, wherein said additive is present as 0.5–5.0 weight percent of the lubricant composition.

13. The lubricant composition of claim 12, wherein said additive is present as 1.0–2.5 weight percent of said lubricant composition.

14. The lubricant composition of claim 11, wherein said lubricating oil is a natural or synthetic hydrocarbonaceous oil.

15. The lubricant composition of claim 11 also containing an effective amount of a succinimide dispersant.

References Cited

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