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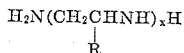
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DETERGENT-DISPERSANT LUBRICANT ADDITIVE HAVING ANTI-RUST AND ANTI-WEAR PROPERTIES

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This invention relates to additives for lubricant oils and more particularly pertains to reaction products of liquid viscous polybutenes, maleic anhydride, tetraethylene pentamine and similar ethylene amines and boric acid. This invention also pertains to the preparation of said reaction products and the use of said reaction products in lubricant oil compositions.

It is known that 3(4) alkyl or alkenyl-substituted succinic anhydrides containing 5 to 20 carbon atoms in the alkyl or alkenyl groups are corrosion inhibitors in lubricating oils and greases. It is also known that monoamine reaction products of such alkyl or alkenyl succinic anhydrides are also corrosion inhibitor additives for lubricating oils and greases. It has become recently known, from French Patent 1,254,094, that mono- and di-amide derivatives of polyamines, such as the "ethylene amines" of the class corresponding to the formula



derived from ethylene (where x is about 1 to about 10) dichloride and ammonia, reacted with aliphatic-substituted succinic acid or anhydride where the aliphatic group contains 50 or more carbon atoms possess detergent properties useful in lubricant oil compositions for internal combustion engines. It has also recently become known, from French Patent 1,265,086, that monosuccinimides of tetraethylene pentamine obtained from 3(4) alkenylsuccinic anhydride having 30 to 200 carbon atoms in the alkenyl group possess detergency properties useful in lubricant oil compositions for internal combustion engines.

The alkylsuccinic anhydride-polyamine detergency derivatives of both the foregoing French patents have several characteristics in common. They both are indicated as having a number of free secondary amino groups and they both contain long chain hydrocarbon substituents in the 3 or 4 position of the succinic anhydride ring moiety. The products of both patents are obtained by reacting long chain hydrocarbons with maleic anhydride at elevated temperatures in the range of 300 to 450° F. in varying ratios of hydrocarbon to anhydride to produce the alkyl or alkenyl succinic anhydride which is then reacted with the polyamine or tetraethylene pentamine at temperatures above 175° F., generally in the range of 200 to 320° F. or higher up to as high as 500° F. with the removal of water of reaction and reaction diluent, such as toluene, if used.

French Patent 1,254,094 characterizes the products obtained by the reaction of the ethylene amines with aliphatic-substituted succinic anhydride or the corresponding acids as reaction products containing simple acyclic diamides, cyclic diimides, or a polymeric amide (the substituted succinic anhydride or acid being an open chain unit of the polyamide). These amide products are said to be formed by the acylation of the polyamine with the aliphatic-substituted succinic anhydrides or acids. Through a reaction differing in kind from said acylation reaction, this French patent suggests that a certain amount of imide formation will occur but does not specify to what extent imide formation occurs. Thus, the reaction mixture of French Patent 1,254,094 is said to contain a mixture of

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undefined acyclic diamides, cyclic diimides, polyamides and perhaps some mono imides. Yet the reaction mixture does, from the data presented, possess detergency properties.

5 French Patent 1,265,086, on the other hand is quite specific with respect to characterizing the hydrocarbon-maleic anhydride-tetraethylene pentamine reaction product as a monoalkenylsuccinimide) of tetraethylene pentamine having three free secondary amino groups and one free primary amino group wherein the alkenyl group substituent on the succinimide ring contains 30 to 200 carbon atoms. These monosuccinimides of tetraethylene pentamine are also lubricant oil detergents as the data demonstrates.

15 The operation of the present day internal combustion engines in automobiles, delivery trucks, taxicabs, police cars, etc. in so-called stop and go city use results in severe demands for high dispersancy and detergency as well as extreme oxidation stability and protection of metal parts to rust and corrosion. This severe demand on lubricant additives comes about by the accumulation of water and partial combustion products in the crankcase of the engines of the foregoing vehicles. Water and many of the partial combustion products result in sludge formation which deposits on the valve train, pistons, piston rings, oil screens and filters. The partial oxidation products, when deposited on hot metal surfaces, form varnishes which impair the efficient operation of the engines. Many solutions to these severe performance demands of city driving conditions have been sought, but while solutions to the dispersing of water, sludge and varnish-forming materials in the lubricating oil was of paramount importance, it was also of paramount importance that the additives to accomplish the dispersing had to be thermally stable at high temperatures of continuous operation under high speed highway and expressway traveling. Also with the advent of the recommendation by internal combustion engine manufacturers of longer intervals between draining and replacement of crankcase lubricants, emphasis has also been directed at the protection of bearings and other metal parts of the internal combustion engines against corrosion, rust and wear. To further complicate the problems of formulating crankcase lubricants for the increasingly severe service demanded by the improved internal combustion engines, there was the always present problem of compatibility of the other additives such as the anti-wear, anti-corrosion, anti-rust and anti-oxidation additives. In many cases by using what was found to be the desired concentration level of the anti-rust, anti-corrosion, anti-oxidation additives with the detergent and/or dispersant, it was found that excessive amounts of these other additives were required by lubricant oil compositions containing the detergent-dispersants derived from the products of reaction between alkenyl succinic anhydride and polyamines of the ethylene amine type. By adjustment of ratios of various additive materials, compatibility could be achieved at the sacrifice of bearing wear or bearing corrosion, rust inhibition and even oxidation stability.

20 We have now discovered that the various reaction products derived from an alkenyl succinic anhydride, a boron compound and the hereinbefore defined ethylene amines, in the manner and sequence hereinafter described, possess not only outstanding detergency-dispersancy properties but also possess anti-corrosion, anti-wear, anti-rust and anti-oxidation properties for use in lubricants for internal combustion engines and at the same time permit the use of lower compatible amounts of anti-wear, anti-corrosion, anti-rust and anti-oxidant additives than would otherwise be required. The aforementioned reaction products are prepared by combining at an elevated temperature a boron compound, e.g. boric acid, with an alkenyl

succinic anhydride to provide an amount of boron suitably from 0.4 to 1.5 moles, desirably 0.6 to 1.3 moles and preferably 0.7 to 1.0 mole of boron per mole of nitrogen in the ethylene amine reactant. Suitably said boron compound and said succinic anhydride are combined at 120 to 400° F. and preferably at 150 to 320° F. This product is then reacted without separate recovery, with an ethylene amine in an amount of from 0.3 to 2.0 moles, preferably 0.4 to 0.7 mole, per mole of alkylene substituted succinic anhydride. More than 1.5 moles boron per mole of nitrogen in the amine reactant causes boric acid to crystallize out of the dispersant on standing, although apparently stable after reaction.

The nature of combining the boron compound, e.g. boric acid with the alkenyl substituted succinic anhydride at elevated temperature is not understood. It may be that boric acid in some way combines with the alkenyl succinic anhydride at temperatures of from 120 to 400° F. and some evidence in other reactions points to such a possibility. However, reaction between alkenyl succinic anhydride and boric acid has not been established with certainty and it is not desired that our invention be based on this theory. It has been established with certainty that the use of up to 1.5 mole of boron per mole of nitrogen in the amine to be later reacted forms a stable final product regardless of the theory of how boric acid and the succinic anhydride reactant combine in the process of this invention. Since the certainty of the overall reactions involved have not been ascertained, the resulting products cannot be defined by conventional terms, e.g. chemical structure, and they are for this reason defined as reaction products of the process from which they are derived.

The improvement provided by this invention is obtained from reaction products resulting from alkenyl substituted succinic anhydrides wherein the alkenyl substituents contain 30 to 200 carbon atoms. Such alkenyl substituents are preferably derived from viscous liquid polyolefins of an average molecular weight in the range of 500 to 100,000 and preferably viscous liquid polybutenes of the foregoing characteristics. These alkenyl substituted succinic anhydrides are combined at elevated temperature with a boron compound such as boric acid, boric acid anhydride, boric acid esters and boron trifluoride among others. Boric acid is the preferred boron compound. It is also preferred that the polyamine of the ethylene amine type from which the final dispersant-detergent is derived is a tetraethylene pentamine, i.e. tetraethylene pentamine per se or the commercially available products consisting primarily of tetraethylene pentamine admixed with related ethylene amines which mixture has an average nitrogen content of tetraethylene pentamine. Thus, the preferred final reaction product is derived by the use of from about 1 to 3.5 moles boric acid with one mole of the alkenyl succinic anhydride and with 0.4 to 0.7 mole of a tetraethylene pentamine per mole of alkenyl succinic anhydride.

For combining with the alkenyl substituted succinic anhydride, boric acid, boric acid anhydride or a boric acid ester, either as a solid or as a solution in a solvent such as dioxane, acetone or methanol or as a suspension in light hydrocarbon oil can be used. There is advantageously employed as an aid for said combination a hydroxylic compound such as water or a lower (C₁ to C₄) alkanol such as methanol, ethanol, propanol, isopropanol, n-butanol, isobutanol and the like, or mixtures of water with any one of these alcohols.

It is preferred to heat the mixture of alkenyl substituted succinic anhydride and boric acid, the latter advantageously initially dispersed in a light mineral oil, such as SAE-5W oil, and at about 120 to 150° F. to about 200° F. for a short period of time, 10 to 60 minutes, then add the polyamine permitting the heat of reaction to increase the temperature of the mixture to about 250° F., add heat to 300 to 380° F. and remove water of reaction, advantageously with nitrogen stripping. The resulting substan-

tially water-free product is filtered, preferably using a filter aid. Filter aids such as diatomaceous earth, Fuller's earth and the like can be used.

As hereinbefore stated, the monoalkenyl succinic anhydride is prepared by reacting an excess of polyolefin, especially polybutene, with maleic anhydride. This excess unreacted polyolefin, especially polybutene is retained with the resulting alkenyl succinic anhydride and carried over into the finished formulated lubricating oil, for the unreacted polyolefin, especially the polybutenes, have no adverse effect on the function of the finished formulated lubricating oil. Also, as hereinbefore stated, a small amount of light hydrocarbon oil, e.g. solvent extracted SAE 5W may also be present to provide suitable viscosity characteristics for further process handling especially when the resulting alkenyl substituted succinic anhydride is further reacted in a different reaction vessel.

By removal of water to a "substantially water-free product" is meant the removal of substantially all of the by-product water to a water content of about 2.0% by weight maximum for such a dried product has enhanced storage stability. It appears that about 2.0% by weight of water in the final reaction product is the maximum which can be tolerated without any deleterious effect on viscosity of the reaction mixture and/or hydrolysis of the reaction product with the associated precipitation of boric acid.

The substantially dry reaction product is filtered hot, that is, at a temperature of 280 to 320° F. to remove any undissolved material. The resulting filtered reaction product is a suitable concentrate for dilution to 0.5 to 5% concentration by weight of reaction product with base oil lubricant stocks such as hydrocarbon lubricant oils and synthetic oils to formulate finished crankcase lubricant compositions of the desired viscosity or viscosity range corresponding for example to a SAE 10, SAE 20, SAE 30 or a SAE 10W-SAE 30 multigrade crankcase lubricating composition. Other lubricant additives can be incorporated to meet the severity of service for which the finished lubricating oil composition is designed.

Before illustrating the preparation and use of the products of this invention, the inventive concept with respect to these products, their preparation and their use is herewith summarized. The multifunctional lubricant additive is a complex reaction product obtained by combining a monoalkenylsuccinic anhydride with a boron compound (e.g. boric acid, boric anhydride or boric acid ester) at 120 to 400° F. to form a first product, and then reacting the first product with an ethylene amine of the type and class hereinbefore defined under temperature conditions to split out water and also to drive off by-product water. The ratio of said ethylene amine to monoalkenyl succinic anhydride-boric compound first product is suitably 0.3 to 2.0 moles, and preferably 0.4 to 0.7 mole per mole of said substituted succinic anhydride. The reaction temperature for the amine with said succinic anhydrideboron compound is in the range of 175 to 500° F. The alkenyl group in said substituted succinic anhydride has 30 to 200 carbon atoms and is preferably derived from liquid viscous polybutene. The amount of boron compound is in the range to provide 0.5 to 1.5 boron per nitrogen in the final reaction product. To insure a haze-free final reaction product, it is preferred to filter and strip the reaction product with an inert gas at 300° F. In any of the final reaction products, the final amount of water is a maximum of 2% by weight of the solution of the reaction product.

The complex reaction product can be prepared as such but it is preferred to prepare it as a solution in a base lubricating oil or as a solution in a polyolefin corresponding to the 30 to 200 carbon atom alkenyl group substituent. In either case it is desirable to use a 40 to 80 weight percent solution of monoalkenyl succinic anhydride. It is also desired to have the final reaction product as a solution containing from about 40 to 60 weight percent of the reaction product in a solvent comprising a lubricant base

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of a viscosity in the range of SAE 5 and SAE 5W weight hydrocarbon oils and unreacted polyolefin to form a concentrate suitable for dilution with other base lubricants and additives to a range of from 0.5 to 5 weight percent complex boration product, preferably 1 to 3 weight percent reaction product to obtain a completely formulated long life, maximum severity service oil.

The preferred complex reaction product is obtained from each mole of mono-(polybutenyl)-succinic anhydride (M.W. of substituent is 860) reacted with about 1 to 3.5 moles boric acid and 0.4 to 0.7 mole of tetraethylene pentamine.

The process and products of this invention can be more clearly understood from illustrative examples of the practice of the process producing representative reaction products and the use of the reaction products of this invention in finished lubricating oil formulations for use in lubricating the crankcase and valve trains of internal combustion engines as hereinafter set forth in specific examples.

In all of the following examples using polybutenyl succinic anhydride reactant, this reactant is obtained by reacting 0.85 mole of maleic anhydride per mole of polybutene having a molecular weight of about 860. Although not all of the maleic anhydride enters into the reaction at about 450° F., only about 5 weight percent is thermally decomposed and a small amount is stripped out. The yield of polybutenyl succinic anhydride is about 58% based on the polybutene used.

Example 1

To 2000 parts by weight of solution of 48.3% polybutenyl succinic anhydride in polybutene of 860 molecular weight heated to 150° F. there is added 140 parts boric acid and 25 parts water both by weight. The resulting mixture is heated to 200° F. with stirring. Then 97 parts tetraethylene pentamine are added with stirring at a rate to increase the temperature of the mixture to about 280° F. by heat of reaction. The resulting reaction mixture is stirred, heated to 350° F. and stripped with nitrogen for about 2.5 hours. Thereafter the mixture is filtered through celite diatomaceous earth filter aid at 300° F. The product after dilution with 177 parts of SAE-5W oil is a dark, clear viscous liquid which by analysis is found to contain 0.9% boron and 1.35% nitrogen.

Example 2

The process of Example 1 is repeated except that 140 parts boric acid and 35 weight parts of methanol are used in place of the 140 parts of boric acid and 25 parts of water by weight. A dark, slightly hazy but otherwise clear viscous liquid product is obtained. The product by analysis is found to contain 0.87% boron and 1.31% nitrogen.

Example 3

To 2000 weight parts of polybutenyl succinic anhydride (48.3% active) at 180–200° F. there is added with stirring a hot dispersion of 158 parts of boric acid in 177 weight parts SAE-5W oil (120–150° F.). The mixture is heated to and stirred at 200° F. for 30 minutes. Thereafter, 97 weight parts of tetraethylene pentamine are added. During the addition of the polyamine the reaction mixture temperature increases to 250–280° F. The reaction mixture is heated to 380° F. and is blown at this temperature for 2.5 hours with a stream of nitrogen. The reaction mixture is then filtered at 300° F. through a Celite diatomaceous earth pad. The product obtained is a dark brown, clear, viscous liquid consisting of 45% of active dispersant-rust inhibitor in base oil.

A series of preparations of reaction products of this invention are conducted using as the reactants: polybutenyl succinic anhydride solution in a solvent consisting of polybutene (860 M.W.) and SAE-5W oil, commercial mixture of polyamines having an average nitrogen content equivalent to tetraethylene pentamine designated "TEPA" hereinafter and boric acid dispersed in hot SAE-

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5W oil (in the ratio of 1.0 lb.-mole of boric acid in 8.5 gallons of oil) are combined under different temperature conditions. The mole ratio of each boric acid and TEPA to polybutenyl succinic anhydride are constant at about 2.2 and 0.5, respectively. Additional SAE-5W oil is added before filtering the final reaction product and is added in an amount to provide a concentration of 45–50 weight percent of the final reaction product. The precise amount added will depend on the concentration of the solution of polybutenyl succinic anhydride used. For example, when a 60% solution of polybutenyl succinic anhydride (molecular weight of 960) is used and boric acid dispersion of 6.8 pounds per gallon of oil to provide 2.5 moles boric acid per mole of polybutenyl succinic anhydride is used followed by reaction of 0.5 mole TEPA per mole of the substituted succinic anhydride, the addition of about 3.5 gallons of SAE-5W oil per mole of the substituted succinic anhydride will produce a 50% concentrate of the final reaction product. The nitrogen blowing time should be from 1–3 hours at 300°–380° F. to insure a dry product.

Example 4

For each mole of polybutenyl succinic anhydride at 250° F. there is added 2.25 moles boric acid suspension in SAE-5W oil (1.0 lb. mole per 8.5 gallons) and the mixture is stirred and heated to 360° F. for about one hour. The resulting product is cooled to 320° F. and tetraethylene pentamine, 0.5 mole per mole of polybutenyl succinic anhydride, is added slowly to the stirred first product so that the temperature of the reaction mixture does not exceed 360° F. during addition of the polyamine. Thereafter, the reaction mixture is heated to 380° F. and by-product water is removed in about 3.0 hours aided by nitrogen stripping. The dried reaction mixture is diluted with SAE-5W oil to about 50% concentration of the final reaction product and then filtered. This filtered concentrate is found by analysis to contain 1.44% nitrogen and 0.9% boron.

Example 5

In this preparation the polybutenyl succinic anhydride solution and the boric acid dispersion are combined in a mole ratio of 1.5 H₃BO₃ per mole of anhydride and stirred at 150° F. After about 10 minutes the TEPA is added with stirring producing a temperature rise to 230° F. The resulting mixture is heated to 300° F. and by-product water is removed in about 2 hours with the aid of nitrogen stripping. This reaction mixture is diluted to a 50% concentrate of the reaction product by the addition of SAE-5W oil and then filtered. This filtered concentrate is found by analysis to contain 1.45% nitrogen and 0.52% boron. This preparation, based on the final nitrogen and boron content of the concentrate is slightly less efficient with respect to the utilization of the boric acid.

Example 6

The preparation of Example 1 is repeated except that 50 parts of water are used. The filtered concentrate (50% reaction product) is found by analysis to contain 1.27% nitrogen and 0.72% boron.

Example 7

In this preparation the polybutenyl succinic anhydride solution and boric acid dispersion in oil are combined and stirred. Then about 1.5 moles of water per mole of boric acid is added. This mixture is heated to 300° F. and held at this temperature for about 2.0 hours before the TEPA is added. The boric acid-substituted succinic anhydride product can be cooled to 250° F. before addition of TEPA or the TEPA can be added at 300° F. under reflux conditions. The latter is used in this preparation. The filtered dried 50% concentrate is found by analysis to contain 1.35% nitrogen and 0.9% boron.

Example 8

In this preparation 770 gallons solution containing 60% by weight polybutenyl succinic anhydride (M.W. 960) at 325° F., 20 pounds of water and 316 pounds (8.35 pound-moles) boric acid in 75 gallons of SAE-5W oil at 70° F. are combined, stirred and heated at 260° F. for about one hour. Then 1.67 pound moles TEPA (316 pounds) are added slowly with stirring over about 90 minutes. The resulting mixture is diluted with 118 gallons of SAE-5W oil at about 250° F. and then heated to 300° F. By-product water is removed with the aid of nitrogen stripping until the reaction mixture concentrate contains less than 1.0% water, 2 to 4 hours. About 25 pounds of filter aid is added, the reaction mixture concentrate is circulated at 300° F. through a Sparkler filter until a passing blotter test is obtained, and then filtered. The concentrate product is collected.

Ingredient:	Weight percent
SAE-10 base oil	94.29
Zinc dialkyldithiophosphate	1.2
Polymeric fatty acid (mixture of dimer-trimer fatty acids)	0.16
Product of Example 9	4.35

The foregoing crankcase lubricating oil formulation is used in the Oldsmobile MS Sequences I, II, III tests which is designed to evaluate the inhibition of engine rusting. Average rust on lifters and of the engine in general is of interest from the results of this test. The rust values are determined visually according to a scale of from 1 to 10 with 10 as perfect (entirely free of rust), and to pass this test a premium oil must have a rating value of at least 8. The Oldsmobile MS Sequences I, II, III are carried out as hereinafter indicated.

OLDSMOBILE MS RUST TEST

	Test Sequence		
	I	II	III
Engine	Oldsmobile		
Operating Conditions:			
Speed, R.p.m.	2,500±20	1,500±20	3,400±20
Load, B.h.p.	None	25±2	85±2
Coolant Out, ° F.	95±2	95±2	200±2
Coolant In, ° F.	85 minimum	85 minimum	190 minimum
Oil Sump, ° F.	120 maximum	120±2	265±2
A/F Ratio		16.0±0.5	16.0±0.5
Intake air humidity, grains/lb. of dry air.	80±5	80±5	80±5
Valve Spring Loading	Normal	Normal	Normal
Oil Consumption, Qts.		5 maximum	
Test Schedule:			
Cycle:			
Operating, Hours	¼ (10 min.)	3	36
Shutdown, Hours	¾ (50 min.) ¹	3	
No. of Cycles per Test	30	16	1
Total Test time, Hours	30	96	36
Oil Filter	No.	No.	No.
Test Fuel:			
Blend		75% Standard Aviation 80 25% Indolene (or equivalent)	
TEL Content, cc./gal.		3.0	
Sulfur, wt. percent		0.16±0.02	
Crankcase Ventilation	Plugged	Plugged	Normal

¹ Cooling water maintained at 95° F. during shutdown.

By the foregoing process an additive concentrate containing about 50% of the final reaction product is obtained which contains about 1.4% nitrogen and 0.9 to 1.0% boron. The concentrate has a typical viscosity of about 800 SSU at 210° F. and a flash point of about 345° F. (Cleveland open cup).

Example 9

To 2,000 grams of 48% solution of polybutenylsuccinic anhydride (polybutenyl M.W. about 860-1.02 moles anhydride by titration) there is added 25 grams of water and 140 grams of boric acid (2.22 moles). This mixture is heated at 150° F. Then there is added 96 grams (0.5 mole) of tetraethylene pentamine over a period of 1.5 hours. The reaction mixture is further heated to 300-320° F. for an hour followed by nitrogen blowing at 300-320° F. for 2-3 hours or until dry. To the reaction mixture there is added 177 grams of SAE-5W base oil. The reaction mixture is filtered at 300° F. through a "Celite" pad. The product obtained is a dark brown viscous liquid consisting of 45% active dispersant-rust inhibitor reaction product. The analyses of the reaction product shows: 1.34% nitrogen and 0.94% boron.

The percent activity of the final reaction product can, of course, be adjusted by varying the amount of oil added. The oil may be added at any point in the procedure previous to filtration. The product of Example 9 is employed in a crankcase lubricating oil formulation containing the following ingredients:

From the foregoing Oldsmobile MS rust tests it was found that the crankcase lubricating formulation containing the reactant product of Example 9 had an average rust rating of 9.2. Representative reaction product of this invention is illustrated by Examples 1 through 8 and can be used within the range of 0.5 to 5% by weight active ingredient in the concentrate. However, for outstanding performance, 2 to 3% of the active ingredient (reaction product) in the reaction product concentrate can be used to formulate a premium lubricating oil having high, stable, long-lasting dispersancy-detergency as well as high anti-rust, anti-wear and oxidation inhibition.

What is claimed is:

1. An oil-soluble lubricant additive which comprises the reaction product of reactants consisting essentially of mono-alkenyl substituted succinic anhydride, a boron compound selected from the class consisting of boric acid and boric anhydride and an ethylene amine having the formula $H_2N(CH_2CH_2NH)_xH$ wherein x is a number from 1 to about 10, wherein said alkenyl group of the substituted succinic anhydrides has from 30 to 200 carbon atoms and said reactants are employed in the mole ratio of 0.3 to 2.0 moles of an ethylene amine per mole of said monoalkenyl succinic anhydride and an amount of boron compound to provide a molar ratio of boron of 0.4 to 1.5 per mole nitrogen in said ethylene amine reactant and said oil-soluble lubricant additive is prepared by combining said substituted succinic anhydride and said boron compound in the proportions to provide the foregoing mole ratios at a temperature in the range of from 120°

to 400° F. to form a first product and then reacting this first product with said ethylene amine in the proportion to provide 0.3 to 2.0 moles of said amine per mole of said substituted succinic anhydride at a temperature in the range of 175 to 500° F. to split out water to obtain a product having a maximum of water not exceeding 2.0%.

2. The product of claim 1 wherein said boron compound is boric acid.

3. The product of claim 1 wherein the monoalkenyl substituted succinic anhydride is derived from polybutene having a molecular weight of about 860, the boron compound is boric acid and said ethylene amine is tetraethylene pentamine.

4. A process of preparing an oil-soluble lubricant additive having only the elements boron, nitrogen, carbon, hydrogen and oxygen and having from 0.4 to 1.5 mole boron per mole nitrogen which comprises combining at a temperature in the range of 120 to 400° F. reactants consisting essentially of a monoalkenyl substituted succinic anhydride whose alkenyl group has 30 to 200 carbon atoms and a boron compound selected from the class consisting of boric acid and boric anhydride to form a first product, reacting said first product at a temperature in the range of 175 to 500° F. with an ethylene amine having the formula $H_2N(CH_2CH_2NH)_xH$ wherein x is a number from 1 to about 10, wherein said reactants are employed in the ratios to provide for each mole of said monoalkenyl substituted succinic anhydride 0.3 to 2.0 moles of said ethylene amine and said boron compound to provide from 0.4 to 1.5 moles boron per mole of nitrogen and removing by-product water to a maximum of 2.0 weight percent based on the reaction product.

5. The process of preparing an oil-soluble lubricant additive from a mono-(polybutenyl)-succinic anhydride whose polybutenyl substituent has a molecular weight of

about 860 present in a hydrocarbon solution in the range of 40 to 80 weight percent, boric acid and tetraethylene pentamine in the ratio of 0.4 to 0.7 mole of said pentamine per mole of said mono-polybutenyl-succinic anhydride and the ratio of boric acid to said pentamine to provide from 0.6 to 1.0 mole boron per mole of nitrogen which process comprises the procedural sequence of first combining said amount of boric acid with the mono-(polybutenyl)-succinic anhydride in the presence of water at 120 to 400° F., thereafter reacting said amount of the pentamine with the substituted succinic anhydride-boric acid product and thereafter heating the reaction mixture to a temperature in the range of 300 to 380° F. to drive off water to provide a product having a maximum of 2.0 weight percent water.

6. A lubricant oil composition comprising a major amount of a lubricant oil and a minor amount sufficient to impart detergency of the product of claim 1.

7. A lubricant oil composition comprising a major amount of a lubricant oil and from 0.5 to 5% by weight of the product of claim 3.

8. A lubricant oil concentrate containing 40 to 80% of the succinic anhydride-boric acid-tetraethylene pentamine reaction product of claim 3 suitable for dilution with lubricant oil base to 0.5 to 5% by weight of said reaction product.

References Cited

UNITED STATES PATENTS

3,087,936 4/1963 Le Suer ----- 252—51.5 X

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