Patented July 11, 1944 2,353,491

UNITED STATES PATENT OFFICE

2,353,491

LUBRICATING OIL COMPOSITION

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No Drawing. Application January 14, 1944, Serial No. 518,260

18 Claims. (Cl. 252—51)

This invention has to do in a general way with mineral oil compositions and is more particularly related to compositions comprised of mineral oil and a minor proportion of an added ingredient which will improve the oil in one or more important respects.

It is well known to those familiar with the art that mineral oil fractions refined for their various uses are in and of themselves usually deficient in one or more respects so that their practical utility is limited even in the particular field for which they have been refined. For example, mineral oil fractions refined for use as lubricants have a tendency to oxidize under conditions of use, with the formation of sludge or acidic oxidation products; also, the lighter fractions such as gasoline and kerosene tend to oxidize with the formation of color bodies, gum, etc. In order to prevent the formation of these products and thereby extend the useful life of the oil fraction, it is common practice to blend with such oil fraction an additive ingredient which will inhibit oxidation, such ingredients being known to the trade as oxidation inhibitors, antioxidants, sludge inhibitors, gum inhibitors, etc.

It is also the practice to add other ingredients to mineral oil fractions for the purpose of improving "oiliness" characteristics and the wear-reducing action of such mineral oils when they are used as lubricants, particularly when the oils are used for the purpose of lubricating metal surfaces which are engaged under extremely high pressures and at high rubbing speeds.

Various other ingredients have been developed for the purpose of depressing the pour point of mineral oil fractions which have been refined for use as lubricants. Most refining treatments provide oils containing a small amount of wax which, without the added ingredient, would tend to crystallize at temperatures which render the oil impracticable for use under low temperature conditions. Additive agents have also been developed for improving the viscosity index of lubricating oil fractions. In the case of internal combustion engines, particularly those operating with high cylinder pressures, there is a decided tendency for the ordinary lubricating oil fractions to form carbonaceous deposits which cause the piston rings to become stuck in their slots and which fill the slots in the oil ring or rings, thus materially reducing the efficiency of the engine. Ingredients have been developed which, when added to the oil, will reduce this natural tendency of the oil to form deposits which interfere with the function of the piston rings.

It has also been discovered that certain types of recently-developed hard metal alloys, such as cadmium-silver alloy bearings, are attacked by ingredients in certain types of oils, particularly oils of high viscosity index obtained by various methods of solvent-refining. This corrosive action on such alloys has led to the development of corrosion-inhibitors which may be used in solvent-refined oils to protect such bearing metals against this corrosive action.

In the lighter mineral oil fractions, such as those used for fuel purposes, particularly in internal combustion engines, it has been found that the combustion characteristics of the fuel may be controlled and improved by adding minor proportions of various improving agents thereto.

The various ingredients which have been developed for use in mineral oil fractions to improve such fractions in the several characteristics enumerated above are largely specific to their particular applications. Therefore, it has been the practice to add a separate ingredient for each of the improvements which is to be effected.

The present invention is predicated upon the discovery of a group or class of oil-soluble reaction products or compounds which, when added to mineral oil fractions in minor proportions, will improve the oil fractions in several respects. Although the present application is directed broadly to all mineral oil compositions, including both lubricating oils and fuel oil distillates, the invention is illustrated herein by the lubricating oil compositions, and for details concerning the preparation and improved properties of the fuel oil compositions contemplated by the present invention, reference is made to copending application Serial No. 427,592, filed January 21, 1942, in which I am a coinventor and of which the present application is a continuation-in-part.

The novel addition agents contemplated by this invention as multifunctional improvers for mineral oils are the condensation products of an aldehyde, an aliphatic polyamine in which each amino group is characterized by the presence of at least one hydrogen atom, and an alkyl-substituted hydroxyaromatic compound which have been further reacted to replace the hydroxyl hydrogen of the condensation product with a metal. Preferred reaction products are those obtained by condensing about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group is characterized by the presence of at least one hydrogen atom and with a quantity of an alkyl-substituted hydroxyaromatic

compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent; and subsequently reacting the condensation product with an alcohol solution of an alkaline earth metal oxide or alkaline earth metal hydroxide to replace the hydroxyl hydrogen of the alkyl-substituted hydroxyaromatic compound.

Aldehydes contemplated by the present invention are the aliphatic aldehydes, representative of which are formaldehyde and acetaldehyde, and the aromatic aldehydes, such as benzaldehyde. Preference, however, is given to the aliphatic aldehydes, formaldehyde being particularly preferred.

The aliphatic polyamines contemplated herein are those in which each amino group is characterized by the presence of at least one hydrogen atom. Such aliphatic polyamines may contain only primary amino groups, only secondary amino groups, or both primary and secondary amino groups. Of this class of amines, preference is given to the diamines in which two primary amino groups are attached to adjacent carbon atoms, and particular preference is given to ethylene diamine. Typical polyamines of the class described by the foregoing are ethylene diamine, propylene diamine and the other members of this homologous series.

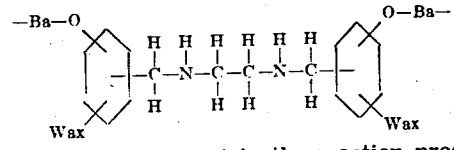
Representative hydroxyaromatic compounds contemplated by the present invention are phenol, resorcinol, hydroquinone, catechol, cresol, xylenol, hydroxydiphenyl, benzylphenol, phenylethylphenol, phenol resins, methylhydroxydiphenyl, guiacol, alpha and beta naphthol, alpha and beta methylnaphthol, tolylnaphthol, xylylnaphthol, benzylnaphthol, anthranol, phenylmethylnaphthol, phenanthrol, monomethyl ether of catechol, methoxyphenol, phenoxyphenol, anisole, beta naphthyl methyl ether, chlorphenol, and the like. Preference in general is to the monohydroxy phenols otherwise unsubstituted, particular preference being given to phenol and alpha and beta naphthol.

As described above, the hydroxyl hydrogen of the alkyl-substituted hydroxyaromatic group in the intermediate condensation product is replaced with its equivalent of metal. Any metal may be used the oxide or hydroxide of which (in alcohol solution, if necessary) can be reacted with the intermediate condensation product, or a salt of which can be reacted in alcohol solution with the sodium salt of the intermediate product. Among the metals contemplated for this purpose are: copper, beryllium, magnesium, calcium, strontium, barium, radium, zinc, cadmium, mercury, germanium, tin, lead, vanadium, chromium, manganese, iron, cobalt, nickel, ruthenium, palladium, platinum, aluminum, antimony, arsenic, bismuth, cerium, columbium, gallium, gold, indium, iridium, molybdenum, osmium, rubidium, selenium, tantalum, tellurium, thorium, titanium, tungsten, uranium, and zirconium. Preference is given to metals of the alkaline earth group, particularly to barium. As noted above, the metal is preferably introduced by reaction of its oxide or hydroxide with the intermediate condensation product. Where necessary, an alcohol solution of the oxide or hydroxide is used. The metal salts can also be prepared by reacting the sodium salt of the intermediate product with an alcohol solution of a salt of the desired metal, such as stannous chloride, lead acetate, thorium nitrate, titanium tetrachloride, etc.

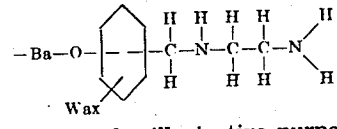
Alcohols suitable for the last-mentioned purposes are methanol, ethanol, etc.

Broadly, the alkyl groups contemplated by the present invention as substituents on the hydroxyaromatic compound are alkyl groups which will impart oil-solubility to the final product. Preferably, such alkyl substituents are long-chain, relatively-high-molecular-weight hydrocarbon groups having at least twenty carbon atoms. Particularly well-adapted for the purpose herein described are alkyl groups derived from petroleum wax, which is a predominantly straight-chain aliphatic hydrocarbon of at least twenty carbon atoms. It will be obvious to those skilled in the art that the maximum number of oil-solubilizing alkyl groups is limited by the number of valences on the aromatic nucleus available for substitution. Naturally, the maximum number of such groups which can be attached to a single aromatic nucleus will vary as the nucleus is mono- or poly-cyclic and as the nucleus is otherwise substituted.

The present application has been purposely directed to reaction products of the aforesaid preferred reactants for as yet the theory of reaction is not fully understood. Some evidence is available—for example, quantitative analysis of the reaction product—to point to the presence of a major quantity of one compound. For instance, when typical reactants, such as a wax-substituted phenol, formaldehyde, ethylene diamine, and barium oxide in alcohol solution, are reacted as hereinafter described, analysis indicates that the predominant product is:



Probably also present in the reaction product is a compound of the following type:



The foregoing is for illustrative purposes only and is not to be construed as limiting the present invention to a theory of reaction because the present invention is directed primarily to reaction products obtained by interreaction of the reactants described herein as multifunctional improving agents for lubricating oils and the like.

As stated above, the general procedure for preparing the contemplated reaction products involves the interreaction of an alkyl-substituted hydroxyaromatic compound, an aliphatic aldehyde, and an aliphatic polyamine wherein the amino groups have at least one free hydrogen, and subsequent reaction or treatment of the product thus obtained to replace or substitute the hydroxyl hydrogen with metal. A typical, and also preferred, alkyl-substituted hydroxyaromatic compound which may be used is a wax-substituted phenol, "wax-phenol." The term "wax" as used herein designates petroleum wax or aliphatic hydrocarbons or hydrocarbon groups of the type which characterize petroleum wax. These so-called "wax" substituents may be obtained by alkylation of the phenol or hydroxy-aromatic hydrocarbon with a relatively-high-molecular-weight aliphatic hydrocarbon or mixture of such

hydrocarbons (such as petroleum wax) by any suitable alkylation procedure, such, for example, as by a Friedel-Crafts condensation of chlorinated petroleum wax with phenol. Furthermore, it is to be understood that the invention is not limited to the use of "wax" or long-chain alkyl groups for imparting oil-solubility but that short-chain or low-molecular-weight alkyl groups which are capable of imparting oil-solubility may be used.

Details of a preferred procedure for making the reaction products of this invention where the aforesaid wax-phenol is employed as the alkyl-substituted hydroxyaromatic compound may be obtained from the following examples:

EXAMPLE 1

A. *Alkylation of phenol*

A paraffin wax melting at approximately 120° F. and predominantly comprised of compounds having at least twenty carbon atoms in their molecules is melted and heated to about 200° F., after which chlorine is bubbled therethrough until the wax has absorbed about 16 per cent of chlorine, such product having an average composition between a monochlor wax and a dichlor wax. Preferably, the chlorination is continued until about one-sixth the weight of the "chlorwax" formed is chlorine. A quantity of chlorwax thus obtained, containing three atomic proportions of chlorine, is heated to a temperature varying from just above its melting point to not over 150° F., and one mol of phenol (C_6H_5OH) is admixed therewith. The mixture is heated to about 150° F., and a quantity of anhydrous aluminum chloride corresponding to about 3 per cent of the weight of chlorwax in the mixture is slowly added with active stirring. The rate of addition of the aluminum chloride should be sufficiently slow to avoid violent foaming, and during such addition the temperature should be held at about 150° F. After the aluminum chloride has been added, the temperature of the mixture may be increased slowly over a period of from 15 to 25 minutes to a temperature of about 250° F. and then should be more slowly increased to about 350° F. To control the evolution of HCl gas the temperature of the mixture is preferably raised from 250° F. to 350° F. at a rate of approximately one degree per minute, the whole heating operation occupying approximately two hours from the time of adding the aluminum chloride. If the emission of HCl gas has not ceased when the final temperature is reached, the mixture may be held at 350° F. for a short time to allow completion of the reaction. However, to avoid possible cracking of the wax, the mixture should not be heated appreciably above 350° F., nor should it be held at that temperature for any extended length of time.

It is important that all unreacted or non-alkylated hydroxyaromatic material (phenol) remaining after the alkylation reaction be removed. Such removal can be effected generally by water-washing, but it is preferable to treat the water-washed product with superheated steam, thereby insuring complete removal of the unreacted material and accomplishing the drying of the product in the same operation.

A wax-substituted phenol prepared according to the above procedure in which a quantity of chlorwax containing three atomic proportions of chlorine and having a chlorine content of 16 per cent is reacted with one mol of phenol will be hereinafter designated as "wax-phenol (3–16)."

B. *Formation of intermediate product*

One hundred atoms of wax-phenol (3–16) prepared according to the foregoing procedure were heated to fluidity, and 12.7 grams of a 69 per cent solution of ethylene diamine in water were added thereto. To give a more fluid mixture about 100 cc. of butanol were also added. To this solution 22.2 grams of a 37 per cent solution of formaldehyde in water were added dropwise, and the reaction was stirred 3½ hours at room temperature and then heated for 7½ hours at a reflux temperature of about 100° C. The reaction mixture was water-washed three times, and the solvent and entrained water were removed by distillation to leave the aliphatic diamino-substituted wax-phenol reaction product.

C. *Metalorganic reaction product*

Twenty-five grams of the product obtained by the procedure described in paragraph B above were blended with 75 grams of mineral oil. Five grams of barium hydroxide octahydrate were added at room temperature. The temperature was slowly raised, and the water of crystallization and the water from the reaction were gradually distilled out. The reaction was held at 150–200° C. for two hours and then cooled. When cool, the reaction mixture was diluted with benzol and filtered through Hi-Flo clay to remove the unreacted barium hydroxide. The diluent was removed by distillation under 20 mm. pressure, and the final product was approximately a ¼ mineral oil blend, which will be hereinafter designated as "Product one."

EXAMPLE 2

A. *Alkylation of phenol*

A paraffin wax melting at approximately 120° F. and predominantly comprised of compounds having at least twenty carbon atoms in their molecules was melted and heated to about 200° F., after which chlorine was bubbled therethrough according to the procedure followed in Example 1, section A, except that the chlorination was continued until the wax had absorbed only about 14 per cent of chlorine. A quantity of chlorwax thus obtained, containing two atomic proportions of chlorine, was heated to a temperature varying from just above its melting point to not over 150° F., and one mol of phenol (C_6H_5OH) was admixed therewith. The mixture was then subjected to the procedure followed in Example 1 to give a wax-substituted phenol which will be hereinafter designated as "wax-phenol (2–14)."

B. *Formation of intermediate product*

One hundred fifty grams of wax-phenol (2–14) prepared according to the foregoing procedure were heated to fluidity, and 20.8 grams of a 69 per cent solution of ethylene diamine in water were added thereto. To give a more fluid mixture, about 100 cc. of butanol were also added. To this solution 19.5 grams of a 37 per cent solution of formaldehyde in water were added dropwise, and the reaction was stirred 3½ hours at room temperature and then heated for 7½ hours at a reflux temperature of about 100° C. The reaction mixture was water-washed three times, and the solvent and entrained water were removed by distillation to leave the aliphatic diamino-substituted wax-phenol reaction product.

C. *Metalorganic reaction product*

Twenty grams of the product obtained by the

procedure described in paragraph B above were blended with 40 grams of mineral oil, and 0.69 gram of sodium as sodium butylate were added thereto. A solution of 3.0 grams of stannous chloride in butanol was gradually added to the reaction mixture at room temperature, and then the reaction was held at the reflux temperature for one hour. The butanol was removed from the reaction mixture by distillation, and the product was filtered through Hi-Flo on a heated funnel to remove precipitated sodium chloride. The final product was approximately a ⅓ mineral oil blend, which will be hereinafter designated as "Product two."

EXAMPLE 3

A. *Alkylation of phenol*

A paraffin wax melting at approximately 120° F. and predominantly comprised of compounds having at least twenty carbon atoms in their molecules was melted and heated to about 200° F., after which chlorine was bubbled therethrough according to the procedure described in Example 1, section A, except that chlorination was continued until the wax had absorbed only about 12 per cent of chlorine. A quantity of chlorwax thus obtained, containing two atomic proportions of chlorine, was heated to a temperature varying from just above its melting point to not over 150° F., and one mol of phenol (C_6H_5OH) was admixed therewith. The mixture was then subjected to the procedure followed in Example 1 to give a wax-substituted phenol which will be hereinafter designated as "wax-phenol (2–12)."

B. *Formation of intermediate product*

One hundred grams of wax-phenol (2–12) prepared according to the foregoing procedure were heated to fluidity, and 7.45 grams of a 60 per cent solution of ethylene diamine in water were added thereto. For greater fluidity about 100 cc. of butanol were also added. To this mixture 12.0 grams of a 37 per cent solution of formaldehyde in water were added dropwise, and the reaction was stirred 3½ hours at room temperature and then heated 7½ hours at a reflux temperature of about 100° C. The reaction mixture was water-washed three times, and the solvent and entrained water were removed by distillation to obtain the final intermediate product, which was a ½ oil blend.

C. *Metalorganic reaction product*

One hundred fifty grams of the product obtained by the procedure described in paragraph B, which as stated above, was a ½ oil blend, were heated to 75° C., and 8.18 grams of barium (as BaO) in methanol were gradually added as the methanol was allowed to distill off. After all the barium oxide in methanol was added, the temperature was raised to 150° C. and held there to remove all of the methanol. The product was filtered through Hi-Flo on a heated funnel, and the final product obtained, which was approximately a ½ oil blend, will be hereinafter designated as "Product three."

EXAMPLE 4

A. *Alkylation of phenol*

A paraffin wax melting at approximately 120° F. and predominantly comprised of compounds having at least twenty carbon atoms in their molecules was subjected to the procedure described in Example 1 except that chlorination of the wax was continued until it had absorbed only about 15 per cent of chlorine and that a quantity of this chlorwax containing only about two atomic proportions of chlorine was admixed with one mol of phenol. This product will be designated hereinafter as "wax-phenol (2–15)."

B. *Formation of intermediate product*

This product was obtained in the same manner as the intermediate product of Example 1 except that the proportions of reactants used were different. Two hundred grams of wax-phenol (2–15) were reacted with 26 grams of a 37 per cent solution of formaldehyde in water and 27.8 grams of a 69 per cent solution of ethylene diamine in water.

C. *Metalorganic reaction product*

The procedure of Example 1 was also followed in this step except that here again the proportions of reactants used were different. Twenty-five grams of the product obtained above were blended with 75 grams of mineral oil, and 10.9 grams of barium hydroxide octahydrate were added thereto. The final product was approximately a ¼ mineral oil blend and will be designated hereinafter as "Product four."

It will be noted in these four examples that the proportions of reactants used in the preparation of the intermediate product were different in each instance, but no difference in the products obtained was noticed as a result of this variation.

As stated hereinbefore, the reaction products contemplated by this invention and illustrated by the above examples, when added to lubricating oils in minor proportions, have been found to improve these oils in several important respects. This phenomenon is demonstrated by the following tables, which give the results of the various tests conducted to determine the effectiveness of these reaction products as addition agents for lubricating oils. The per cent of material added to the oil in the following tables is the per cent of concentrated material and does not include the oil in which the product was made.

POUR POINT DEPRESSION

Tests were conducted in the conventional manner to determine the A. S. T. M. pour points of blends of these reaction products with a Mid-Continent solvent-refined oil of Saybolt Unversal viscosity of 67 seconds at 210° F. as compared with the pour point of the blank oil. The results given in Table I below demonstrate the effectiveness of the reaction products contemplated herein as pour point depressants.

Table I

Addition agent	Per cent added	A. S. T. M. pour point values	
		Inhibited oil	Uninhibited oil
		°F.	°F.
Product one	⅛	−20	+20
Product two	⅛	−20	+25
Product three	1/16	−5	+20
Product four	⅛	−20	+25

VISCOSITY INDEX IMPROVEMENT

A mineral oil of 41.8 seconds Saybolt Universal viscosity at 210° F. was tested in the conventional manner to determine the improvement in viscosity index values effected by various of the reaction products contemplated by this invention. This improvement is clearly demonstrated by the results set forth in Table II below.

Table II

Addition agent	Per cent added	Viscosity index values	
		Inhibited oil	Uninhibited oil
Product one	1	90.7	77.7
Product two	1	84.8	77.7
Product four	1	89.4	77.7

CORROSION TEST

In this test the reaction product was blended with a Pennsylvania solvent-refined oil of Saybolt Universal viscosity of 53 seconds at 210° F., and a section of a bearing containing a cadmium-silver alloy surface and weighing about 6 grams was added to this blend. The oil was heated to 175° C. for 22 hours while a stream of air was bubbled against the surface of the bearing. The loss in weight of the bearing during this treatment measured the amount of corrosion that had taken place. A sample of the straight oil was subjected to the same test at the same time, and the difference between the losses in weight of the two bearing sections demonstrated conclusively the effectiveness of the reaction products contemplated herein as corrosion-inhibitors.

Table III

Addition agent	Per cent added	Bearing loss (weight in mgms.)	
		Inhibited oil	Uninhibited oil
Product four	1/2	1	35

SOCONY-VACUUM TURBINE TEST

Twenty-five cc. samples of a furfural-refined Rodessa crude of Saybolt Universal viscosity of 150 seconds at 100° F. and of a blend of this same oil and Product two were subjected to the following test to determine the effectiveness of the reaction products contemplated by this invention as inhibitors for turbine oils: To each sample were added 1 gram of iron granules and 24 inches of 18 gauge copper wire. The samples were then heated to a temperature of 200° F. with 5 liters of air per hour bubbling therethrough. Two cc. of distilled water were added each day. The results of the tests which were made for color and acidity or neutralization number and amount of sludge formed after certain time intervals are set forth in Table IV below.

Table IV

Inhibitor	Per cent added	Hours	Lovibond color	N.N.	Sludge in mgms.
Product two	1/4	167	4	.01	14
		384	137	23.0	127
None		162	245	17.8	389

It will be noted that although 389 mgms. of sludge had been formed in the uninhibited oil at the end of only 162 hours, only 14 mgms. had been formed in the oil blend at the end of substantially the same length of time and only 127 mgms. had been formed in the inhibited oil at the end of 384 hours.

CFR RING-STICKING TEST

This test was conducted with an oil of Saybolt Universal viscosity of 120 seconds at 210° F. and was carried out in a single-cylinder CFR engine cooled with a diethylene glycol-water mixture held at a temperature of about 390° F. The engine was operated continuously over a time interval of twenty-eight hours at a speed of about 1200 R. P. M., which is equivalent to a road speed of about 25 miles per hour. The oil temperature was held at about 150° F. during the test.

The conditions observed at the end of the test were: the extent to which the piston rings were stuck; the extent to which the slots in the oil rings were filled with deposit; the amount of carbonaceous deposits in the oil; and the neutralization number (N. N.) or acidity of the oil at the end of the test. Comparative runs were made with the oil alone and with the oil containing one per cent of Product three. Results of these runs are given in Table V below.

Table V

Inhibitor	Per cent added	Ring-sticking, in degrees					Per cent slots filled			Total deposits	N. N.
		1	2	3	4	5	1	2	3		
None		360	360	360	360	360	90	80	80	18.0	2.6
Product three	1	0	0	360	90	(1)	30	20	5	6.8	1.8

[1] Tacky.

It will be apparent from the foregoing data that the reaction products contemplated herein are effective not only to inhibit corrosion and the various effects of oxidation upon lubricating oils, such as formation of sludge and color bodies and other undesirable products, but also to depress the pour point and improve the viscosity index of such oils.

The improved properties obtained and the degree of improvement effected may be varied with the aldehyde, metal, polyamine, and aromatic constituents in the reaction products, as well as by the degree of alkylation of the hydroxyaromatic nucleus. As to the degree of alkylation, it is important that the hydroxyaromatic nucleus be sufficiently alkylated to provide a final reaction product "soluble" or "miscible" in the particular lubricating oil fraction with which it is to be blended: that is, a reaction product which will remain uniformly dispersed in the oil in sufficient amount to effect the desired improvement. The amount of improving agent to be used varies with the mineral oil or lubricating oil fraction with which it is blended and with the properties desired in the final oil composition. These reaction products may be added to mineral oils in amounts of from about .05 per cent to about 10 percent, but amounts of from about 0.5 per cent to about 3 per cent are preferred for the purposes of this invention.

It is to be understood that although I have described certain preferred procedures which may be followed in the preparation of the novel reaction products contemplated herein as multifunctional addition agents for mineral oils and have indicated representative reactants for use in their preparation, such procedures and reactants are merely illustrative and the invention is not to be considered as limited thereto or thereby but includes within its scope such changes and modifications as fairly come within the spirit of the appended claims.

I claim:

1. An improved mineral oil composition com-

prising a mineral oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting an aliphatic aldehyde, an aliphatic polyamine in which each amino group has at least one hydrogen atom, and an alkyl-substituted hydroxyaromatic compound wherein the alkyl substituent has a sufficient chain length to impart oil-solubility to the final product; and then substituting the hydroxyl hydrogen in the condensation product thus obtained with metal.

2. An improved mineral oil composition comprising a mineral oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a sufficiency of an alkyl-substituted hydroxyaromatic compound such that the content of the hydroxyaromatic radical in the reaction mixture is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

3. A mineral oil composition comprising a mineral oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting an aldehyde, an aliphatic polyamine in which each amino group has at least one hydrogen atom, and an alkyl-substituted hydroxyaromatic compound wherein the alkyl substituent has a sufficient chain length to impart oil-solubility to the final product; and then substituting the hydroxyl hydrogen of the condensation product thus obtained with metal.

4. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical in the reaction mixture is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with a compound selected from the group consisting of metal oxides and metal hydroxides, said compound containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

5. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical in the reaction mixture is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; substituting the hydroxyl hydrogen in the condensation product thus obtained with sodium to form the sodium salt; and then reacting said sodium salt with an alcohol solution of another metal salt to form the corresponding salt of said last-mentioned metal.

6. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of formaldehydre with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

7. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of alkaline-earth oxides and alkaline-earth hydroxides, said alcohol solution containing an equivalent of metal sufficient to replace the hydroxyl hydrogen of the aforesaid condensation product.

8. An improved mineral oil composition comprising a mineral oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting an aliphatic aldehyde, an aliphatic polyamine in which each amino group has at least one hydrogen atom, and an alkyl-substituted hydroxyaromatic compound wherein the alkyl substituent has a sufficient chain length to impart oil-solubility to the final product; and then substituting the hydroxyl hydrogen in the condensation product thus obtained with barium.

9. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom, and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the

alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of barium oxide and barium hydroxide, said alcohol solution containing an equivalent of barium to replace the hydroxyl hydrogen of the aforesaid condensation product.

10. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom, and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with a compound selected from the group consisting of barium oxide and barium hydroxide, said compound containing an equivalent of barium to replace the hydroxyl hydrogen of the aforesaid condensation product.

11. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic diamine in which each amino group has at least one hydrogen atom, and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

12. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic diamine having two primary amino groups attached to adjacent carbon atoms, and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

13. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic diamine in which each of the two amino groups has at least one hydrogen atom and the amino groups are attached to adjacent carbon atoms, and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

14. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of ethylene diamine and with a quantity of an alkyl-substituted hydroxyaromatic compound sufficient that the content of the hydroxyaromatic radical is about one mol equivalent, the alkyl substituent in the last-mentioned compound having a sufficient chain length to impart oil-solubility to the final product; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

15. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom and with a hydroxyaromatic compound substituted with at least one oil-solubilizing alkyl group having at least twenty carbon atoms, the said substituted hydroxyaromatic compound being present in an amount such that the content of the hydroxyaromatic radical in the reaction mixture is about one mol equivalent; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

16. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom, and with a phenol substituted with a wax-alkyl group, said phenol being present in an amount such that the content of the said phenol in the reaction mixture is about one mol equivalent; and then reacting the condensation product obtained by the foregoing procedure with an alcohol solution of a compound selected from the group consisting of metal oxides and metal hydroxides, said alcohol solution

containing an equivalent of metal to replace the hydroxyl hydrogen of the aforesaid condensation product.

17. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom, and with a hydroxyaromatic compound substituted with at least one oil-solubilizing alkyl group having at least twenty carbon atoms, the said substituted hydroxy-aromatic compound being present in an amount such that the content of the hydroxyaromatic radical in the reaction mixture is about one mol equivalent; and then substituting the hydroxyl hydrogen in the condensation product thus obtained with metal.

18. An improved lubricant composition comprising a lubricating oil having admixed therewith a minor proportion of a reaction product obtained by first interreacting about one mol equivalent of an aliphatic aldehyde with at least one-half mol equivalent of an aliphatic polyamine in which each amino group has at least one hydrogen atom, and with a phenol substituted with a wax-alkyl group, said phenol being present in an amount such that the content of the said phenol in the reaction mixture is about one mol equivalent; and then substituting the hydroxyl hydrogen in the condensation product thus obtained with metal.

EDWARD A. OBERRIGHT.

CERTIFICATE OF CORRECTION.

Patent No. 2,353,491. July 11, 1944.

EDWARD A. OBERRIGHT.

It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction as follows: Page 3, second column, line 2, for "atoms" read --grams--; and that the said Letters Patent should be read with this correction therein that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 16th day of January, A. D. 1945.

Leslie Frazer

(Seal) Acting Commissioner of Patents.