Patented Jan. 11, 1949 2,458,527

UNITED STATES PATENT OFFICE

2,458,527

MINERAL OIL COMPOSITION

Edward A. Oberright, Woodbury, N. J., assignor to Socony-Vacuum Oil Company, Incorporated, a corporation of New York

No Drawing. Application July 6, 1945, Serial No. 603,588

18 Claims. (Cl. 252—33.6)

1

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

2

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 condensation products or compounds which, when added to mineral oil fractions in minor proportions, will improve the oil fractions in several respects.

The novel addition agents contemplated by this invention as multi-functional improvers for mineral oils are the oil-soluble condensation products obtained by reaction of an aldehyde, a polyamine in which each amino group is characterized by the presence of at least one hydrogen atom, and a hydroxyaromatic carboxylic acid (and the metal salts thereof). Preferred condensation products, and metal salts thereof, are those obtained by condensing about one molar equivalent of an aldehyde with at least one-half molar equivalent of a polyamine of the aforementioned character and with a quantity of an alkyl-substituted hydroxyaromatic carboxylic acid sufficient that the content of the hydroxyaromatic substituent is about one molar equivalent. The condensation is preferably carried out in the presence of a suitable solvent.

Aldehydes contemplated by the present invention are the aliphatic aldehydes typified by formaldehyde (such as trioxymethylene), acetaldehyde and aldol (B-hydroxy butyraldehyde); aromatic aldehydes, representative of which is benzaldehyde; heterocyclic aldehydes, such as furfural, etc. The aldehyde may contain a substituent group such as hydroxyl, halogen, nitro and the like; in short, any substituent which does not take a major part in the reaction. Preference, however, is given to the aliphatic aldehydes, formaldehyde being particularly preferred.

The polyamines contemplated herein are those in which each amino group is characterized by the presence of at least one hydrogen atom. Such polyamines may contain only primary amino groups, only secondary amino groups, or both primary and secondary groups. Typical polyamines are the aliphatic homologs, ethylene diamine, propylene diamine, polyalkene polyamines (e. g., diethylene triamine, triethylene tetramine); the aromatic homologs, o-, m- and p-phenylene diamine, diamino naphthalenes, etc. 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 accorded ethylene diamine.

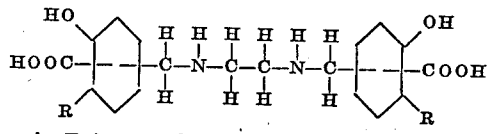
Representative hydroxyaromatic carboxylic acids contemplated herein are phenol carboxylic acids, such as salicyclic acid, naphthol carboxylic acids, etc. Phenol carboxylic acids, however, are preferred.

As indicated hereinabove, the hydroxyaromatic carboxylic acid may contain one or more alkyl substituents, such as short-chain groups, typified by methyl, ethyl, amyl, etc., or long-chain, relatively high molecular weight hydrocarbon groups having at least twenty carbon atoms, typified by alkyl groups derived from petroleum wax, which is a predominantly straight-chain aliphatic hydrocarbon of at least twenty carbon atoms.

A typical, and also preferred, alkyl-substituted hydroxyaromatic carboxylic acid which may be used is a wax-substituted phenol carboxylic acid. 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 a hydroxyaromatic compound 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 to form "wax-phenol," followed by a carboxylation of the "wax-phenol."

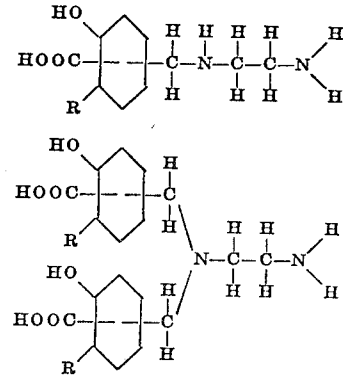
It will be obvious to those skilled in the art that the maximum number of oil-solubilizing alkyl groups is limited by the number of valences on the aromatic nucleus available for substitution. Naturally, the maximum number of such groups which can be attached to a single aromatic nucleus will vary as the nucleus is mono- or poly-cyclic and as the nucleus is otherwise substituted, with such groups as nitro, amino, halogen and the like.

The present application has been purposely directed to condensation products of the aforesaid reactants for as yet, the theory of reaction is not fully understood. Some evidence is available—for example, quantitative analysis of the reaction products—to point to the presence of a major quantity of one compound. For instance, when typical reactants, such as an alkyl-substituted phenol carboxylic acid, formaldehyde and ethylene diamine, are reacted as hereinafter described, analyses indicate that the predominant product is:

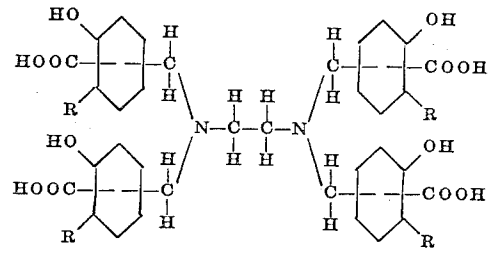


wherein R is an alkyl group.

Probably also present in the reaction product are compounds of the following type:



and



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 invention is directed primarily to condensation products obtained by interreaction of the reactants described herein as multifunctional improving agents for lubricating oils and the like.

In preparing the condensation products contemplated herein, the reactants may be added to each other in any order. A typical procedure involves adding the aldehyde to an alcohol solution of the hydroxyaromatic carboxylic acid and the amine. The reaction may also be carried out in the presence of other diluents or solvents such, for example, as benzol, chlorobenzene, tetrachloroethane, mineral oil, etc. In the event that mineral oil is used as a diluent, the mineral oil may be retained, rather than separated from the condensation product, thereby providing a mineral oil concentrate. Oil concentrates containing the metal salts of these condensation products are similarly obtained.

The reaction temperature may be varied considerably, depending upon the time of reaction, the specific reactants used, etc. For example, the reaction can be carried out at room temperature (20–25° C.) over a relatively long period of time or at the reflux temperature of the reaction mixture over a comparatively short period. By way of illustration, the reactants, in quantities such as shown in the following example, may be thoroughly mixed at room temperature for several hours and the reaction completed at the reflux temperature of the solvent for an additional period of several hours.

The condensation product may be water washed to assure complete removal of any unreacted amine and this is recommended when the amine is high boiling. When an alcohol is used as a diluent in the reaction, it is distilled from the reaction mixture, thereby also removing any unreacted amine and water of reaction or water added with the reactants (formaldehyde, for example, is generally used in a 37% aqueous solution).

As indicated hereinabove, metal salts of the aforesaid condensation products are also contemplated herein. These salts may contain metal replacing only the hydroxyl hydrogen or only the carboxyl hydrogen of the condensation product, or replacing both the hydroxyl and carboxyl hydrogens thereof. 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.

Details of a preferred procedure for making the condensation products of this invention where the aforesaid "wax-phenol" carboxylic acid is employed as the alkyl-substituted hydroxyaromatic carboxylic acid 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 14% 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% 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 14% is reacted with one mol of phenol will be hereinafter designated as "wax-phenol (3–14)."

B. Wax-Phenol Carboxylic Acid

Two hundred and sixteen grams of sodium butylate in butyl alcohol were added to 783 grams of "wax-phenol (3–14)" in 1566 grams of mineral oil (S. U. V. of 65 seconds at 210° F.) at about 25° C. The temperature was then raised to about 200° C. and CO_2 was bubbled into the reaction mixture for several hours. The resulting mixture was dissolved in benzol and neutralized with aqueous HCl followed by water-washing until the washings were neutral. Benzol was removed by distillation of the water-washed product. The benzol-free product is "wax-phenol" carboxylic acid (3–14).

C. Condensation Product

Three hundred grams of "wax-phenol" carboxylic acid (3–14), a 1:2 mineral oil blend obtained in B above, and 11.1 grams of 69% solution of ethylene diamine in water were dissolved in a butyl alcohol and benzol mixture. To the resulting solution, 10.4 grams of a 37% solution of formaldehyde in water-Formalin were added dropwise at room temperature, about 25° C. The resulting mixture was stirred for three hours at 25° C., and then stirred and heated at reflux temperature, about 110° C., for sixteen hours. The reaction product thus obtained was water-washed, and solvents (butyl alcohol and benzol) were removed from the water-washed product by distilling the latter to 175° C. at a pressure of 10 mms. Entrained water is also removed from the product in this distillation. The reaction product —Product One—thus obtained is a 1:2 oil blend containing 0.71% nitrogen.

EXAMPLE 2

Stannous Salt

Three hundred grams of the disodium salt of "wax-phenol" carboxylic acid (3–14), in a 1:2 mineral oil blend (mineral oil of S. U. V. of 65 seconds at 210° F.) and prepared as described in U. S. Patent No. 2,197,834, was dissolved in benzol. A quantity, 7.5 grams, of a 95% solution of ethylene diamine in water was added to the benzol solution, followed by 9.5 grams of Formalin (37%). The resulting reaction mixture was refluxed at 80° C. for four hours. Thereafter, water was distilled therefrom by heating to 120° C. Stannous chloride (22.2 grams), in butyl alcohol, was then added and the reaction mixture was

refluxed at 110° C. for one hour. Butyl alcohol was removed from the reaction mixture by distilling the latter to a maximum temperature of 200° C. The alcohol-free reaction mixture was then cooled. A small quantity of benzol was added and the benzol-containing mixture was filtered through "Hi-Flo" clay. The filtrate thus obtained was distilled to a maximum temperature of 175° C. at 10 mms. pressure, thereby removing benzol. The product—Product Two—is a 1:2 oil blend containing 0.82% nitrogen and 3.86% tin.

As stated hereinabove, the condensation products contemplated by this invention and illustrated by the foregoing examples, when added to mineral oils in minor proportions, have been found to improve these oils in several important respects. This phenomenon is demonstrated in the following tables, which contain 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 table 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 S. U. V. 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	−10	+25

Viscosity Index Improvement

A mineral oil of 41.8 seconds S. U. V. 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	83.0	77.7
Product Two	1	84.3	74.7

Corrosion Test

In this test the reaction product was blended with a Pennsylvania solvent-refined oil of S. U. V. 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, mgms.	
		Inhibited Oil	Uninhibited Oil
Product One	1/10	2	33

Operation Test

To demonstrate the effectiveness of the condensation products under actual operating conditions of an automotive engine, unblended oils and improved oils, containing the reaction products, were subjected to the Lauson engine test. The tests were carried out in a single-cylinder Lauson engine operated continuously over a time interval of sixteen hours with the cooling medium held at a temperature of about 212° F., and the oil temperature held at about 280° F. The engine was operated at a speed of about 1830 R. P. M. At the end of each test the oil was tested for acidity (N. N.) and viscosity. The base oil used here is a solvent-refined oil having an S. U. V. of 44 seconds at 210° F.

Table IV

Addition Agent	Per Cent Added	S. U. V. @ 210° F.	N. N.
None		55.10	5.2
Product One	½	50.34	2.9

It will be apparent from the foregoing test data that the condensation products, and the metal salts thereof, of this invention are effective multifunctional oil improving agents.

The improved properties obtained and the degree of improvement effected may be varied with the aldehyde, polyamine, hydroxyaromatic carboxylic acid and metal used in their preparation and present in the condensation product. For example, when the hydroxyaromatic carboxylic acid contains one or more of the preferred "wax" substituents, the condensation product obtained therewith is characterized by outstanding pour depressant action.

The amount of improving agent used varies with the mineral oil fraction with which it is blended and with the properties desired in the final oil composition. These condensation products may be added to mineral oil in amounts of from about 0.001% to about 10%, but amounts of from about 0.1% to about 3% generally provide satisfactory improvement.

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 containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of the aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

2. An improved mineral oil containing a minor proportion, from about 0.001 per cent to about 10 per cent, of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of the aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

3. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aliphatic aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aliphatic aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

4. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of formaldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of formaldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

5. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and an aliphatic polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the aliphatic polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

6. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and an aliphatic amine having two primary amino groups attached to adjacent carbon atoms, at a temperature falling within the range varying beteen about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the aliphatic amine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

7. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and ethylene diamine, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the ethylene diamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

8. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, an alkyl-substituted hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the alkyl-substituted hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

9. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a wax-substituted hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the wax-substituted hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

10. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, wax-substituted phenol carboxylic acid and a polyamine in which each amino group has

at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the wax-substituted phenol carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

11. An improved mineral oil containing a minor proportion of an oil-soluble, nitrogen-containing condensation product obtained by reaction of formaldehyde, wax-substituted phenol carboxylic acid and ethylene diamine, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of formaldehyde, being at least about one-half molar equivalent of the ethylene diamine and a sufficient amount of the wax-substituted phenol carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

12. An improved mineral oil containing a minor proportion of an oil-soluble polyvalent metal salt of a nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

13. An improved mineral oil containing a minor proportion, from about 0.001 per cent to about 10 per cent of an oil-soluble polyvalent metal salt of a nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

14. An improved mineral oil containing a minor proportion of an oil-soluble polyvalent metal salt of a nitrogen-containing condensation product obtained by reaction of formaldehyde, wax-substituted phenol carboxylic acid and ethylene diamine, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of formaldehyde, being at least about one-half molar equivalent of the ethylene diamine and a sufficient amount of the wax-substituted phenol carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

15. An improved mineral oil containing a minor proportion of a tin salt of a condensation product obtained by reaction of formaldehyde, wax-substituted phenol carboxylic acid and ethylene diamine, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of formaldehyde, being at least about one-half molar equivalent of the ethylene diamine and a sufficient amount of the wax-substituted phenol carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

16. A mineral oil concentrate containing upwards of 10% of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

17. A mineral oil concentrate containing upwards of 10% of a polyvalent metal salt of an oil-soluble, nitrogen-containing condensation product obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture, and for a time sufficient to effect the condensation reaction; the ratio of reactants used, on the basis of one molar equivalent of aldehyde, being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

18. An improved mineral oil containing a minor proportion of a material selected from the group consisting of an oil-soluble, nitrogen-containing condensation product and a polyvalent metal salt of said condensation product: said product being obtained by reaction of an aldehyde, a hydroxyaromatic carboxylic acid and a polyamine in which each amino group has at least one hydrogen atom, at a temperature falling within the range varying between about 20° C. and the reflux temperature of the reaction mixture and for a time sufficient to effect the condensation reaction; the ratio of reactants used on the basis of one molar equivalent of the aldehyde being at least about one-half molar equivalent of the polyamine and a sufficient amount of the hydroxyaromatic carboxylic acid to provide about one molar equivalent of the hydroxyaromatic substituent thereof.

EDWARD A. OBERRIGHT.

(No references cited.)