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STABILIZATION OF ORGANIC COMPOUNDS

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This application is a continuation-in-part of my copending application Serial No. 647,190, filed March 20, 1957, now abandoned, and relates to the stabilization of organic compounds and more particularly to a novel process for preventing deterioration of said organic compounds in storage, during transportation and/or in use. The novel additive of the present invention is a new composition of matter and also is being so claimed in the present application.

The present invention is particularly advantageous for use in the stabilization of a hydrocarbon distillate and serves to improve the hydrocarbon distillate in a number of different ways. For example, in fuel oils, burner oils, range oils, disel oils, marine oils, turbine oils, cutting oils, rolling oils, soluble oils, drawing oils, slushing oils, slushing greases, lubricating oils, lubricating greases, fingerprint removers, etc., the distillate or grease is improved in one or more ways including retarding and/or preventing sediment formation, dispersion of sediment when formed, preventing and/or retarding discoloration, oxidation inhibitor, rust or corrosion inhibitor, detergent, etc. In lubricating type oils, in addition to all or some of the properties hereinbefore set forth, the additive may function as a pour point depressant, viscosity index improver, antifoaming agent, etc. In liquefied petroleum gases, gasoline, naphtha, aromatic solvents, kerosene, jet fuels etc., the additive serves as a corrosion inhibitor along with one or more of the other functions mentioned above. In gasoline the product also functions as anti or deicing additive, carburetor detergent, etc. In other organic compounds, including alcohols, ethers, chlorinated hydrocarbons, synthetic di- and polyesters such as dioctyl sebacate, dioctyl adipate, trimethylolpropane tripelargonate, trimethylolpropane tricaproate, trimethylolpropane tricaprylate, etc., and compositions containing them, glyceridic oils and fats, waxes, other oils and fats of animal or vegetable origin, etc., the additive functions as a beneficent in one or more of the manners herein set forth or otherwise.

The invention is particularly applicable to the stabilization of hydrocarbon distillates heavier than gasoline. The hydrocarbon distillate may be cracked, straight run or mixtures thereof. Many burner oils and particularly blends of straight run and cracked fuel oils undergo deterioration in storage, resulting in the formation of sediment, discoloration, etc. The formation of sediment is objectionable because the sediment tends to plug burner tips, injectors, etc. In jet fuels, oil-fuel heat exchangers and burner nozzles are plugged. In disel fuel, the deterioration tends to form varnish and sludge in the diesel engine. Discoloration of burner oils is objectionable for various reasons, including customer's preference for light colored oils.

In handling of hydrocarbon distillates and other organic liquids, it is often necessary to transport and/or store such materials in metal containers, as in steel or other metal pipe lines, drums, tanks, etc. Since these materials often contain varying amounts of water in solution or in suspension which may separate, due to temperature changes, internal corrosion of the container by separating water almost invariably occurs to a greater or lesser degree. The water thus separated forms as a film or in minute droplets in the pipe line or on the container walls or even in small pools at the bottom of the container. This brings about ideal conditions for corrosion and consequent damage to the metal surfaces of the container, as well as the serious contamination of the hydrocarbon oil or other materials contained therein by the corrosion products.

Corrosion problems also occur, for example, in the lubrication of internal combustion engines or steam engines, including turbines and other similar machinery, in which a quantity of water is often observed as a separate phase within the lubricating system as a result of the condensation of water from the atmosphere or, in the case of internal combustion engines, as the result of dispersion or absorption in lubricating oil of water formed as a product of fuel combustion. Water in such instances corrodes the various metal parts of the machinery with which it comes into contact, the corrosion products causing further mechanical damage to bearing surfaces and the like due to their abrasive nature and catalytically promoting the chemical degradation of the lubricant. Corrosion problems also arise in the preparation, transportion and use of various coating compositions such as greases, household oils, paints, lacquer, etc., which often are applied to metal surfaces for protective purposes.

In one embodiment, the present invention relates to a process for stabilizing an organic substrate against deterioration which comprises incorporating therein a stabilizing concentration of the reaction product of 1,3-diamino-2-propanol with an aliphatic ketone having at least 8 carbon atoms, preferably at least 12 carbon atoms and more particularly from about 8 to about 12 carbon atoms to about 50 carbon atoms per molecule.

In a specific embodiment the present invention relates to the stabilization of burner oil or jet fuel which comprises incorporating therein a stabilizing concentration of the reaction product of 1,3-diamino-2-propanol with methyl heptadecyl ketone.

In another embodiment, the present invention relates to an organic substrate and particularly burner oil or jet fuel subject to deterioration containing a stabilizing concentration of the reaction product herein set forth.

As will be shown by the data in the appended examples, the reaction product of 1,3-diamino-2-propanol with an aliphatic ketone having at least 8 carbon atoms possesses unexpected and superior properties for use in the stabilization of organic substrates and especially in the stabilization of hydrocarbon oils. The peculiar properties of these reaction products are surprising when compared with the reaction products prepared from cyclic ketones or various other amino compounds. Normally it would be expected that the reaction products of these other amino compounds with ketones or those prepared from cyclic ketones would possess substantially equivalent properties as inhibitors, and it is surprising to find that the reaction products prepared from 1,3-diamino-2-propanol and aliphatic ketones of more than 8 carbon atoms possess the considerably superior properties as illustrated in the examples of the present application.

As hereinbefore set forth, the additive for use in the present invention is prepared by the reaction of 1,3-diamino-2-propanol with an aliphatic ketone having at least 8 and preferably at least 12 carbon atoms. It is essential that the ketone contains at least 8 carbon atoms and preferably at least 12 carbon atoms and usually the ketone will contain from about 8 and preferably from about 12 to about 50 carbon atoms per molecule.

Any suitable aliphatic ketone may be employed. Illustrative ketones containing at least 12 carbon atoms include methyl decyl ketone, methyl undecyl ketone, methyl dodecyl ketone, methyl tridecyl ketone, methyl tetradecyl ketone, methyl pentadecyl ketone, methyl hexadecyl ketone, methyl heptadecyl ketone, methyl

octadecyl ketone, methyl nonadecyl ketone, methyl eicosyl ketone, methyl heneicosyl ketone, methyl docosyl ketone, methyl tricosyl ketone, methyl tetracosyl ketone, methyl pentacosyl ketone, methyl hexacosyl ketone, methyl heptacosyl ketone, methyl octacosyl ketone, methyl nonacosyl ketone, methyl triacontyl ketone, etc., ethyl nonyl ketone, ethyl decyl ketone, ethyl undecyl ketone, ethyl dodecyl ketone, ethyl tridecyl ketone, ethyl tetradecyl ketone, ethyl pentadecyl ketone, ethyl hexadecyl ketone, ethyl heptadecyl ketone, ethyl octadecyl ketone, ethyl nonadecyl ketone, ethyl eicosyl ketone, ethyl heneicosyl ketone, ethyl docosyl ketone, ethyl tricosyl ketone, ethyl tetracosyl ketone, ethyl pentacosyl ketone, ethyl hexacosyl ketone, ethyl heptacosyl ketone, ethyl octacosyl ketone, ethyl nonacosyl ketone, ethyl triacontyl ketone, etc., propyl octyl ketone, propyl nonyl ketone, propyl decyl ketone, propyl undecyl ketone, propyl dodecyl ketone, propyl tridecyl ketone, propyl tetradecyl ketone, propyl pentadecyl ketone, propyl hexadecyl ketone, propyl heptadecyl ketone, propyl octadecyl ketone, propyl nonadecyl ketone, propyl eicosyl ketone, propyl heneiscosyl ketone, propyl doscosyl ketone, propyl tricosyl ketone, propyl tetracosyl ketone, propyl pentacosyl ketone, propyl hexacosyl ketone, propyl heptacosyl ketone, propyl octacosyl ketone, propyl nonacosyl ketone, propyl triacontyl ketone, etc., butyl heptyl ketone, butyl octyl ketone, butyl nonyl ketone, butyl decyl ketone, butyl undecyl ketone, butyl dodecyl ketone, butyl tridecyl ketone, butyl tetradecyl ketone, butyl pentadecyl ketone, butyl hexadecyl ketone, butyl heptadecyl ketone, butyl octadecyl ketone, butyl nonadecyl ketone, butyl eicosyl ketone, butyl heneicosyl ketone, butyl docosyl ketone, butyl tricosyl ketone, butyl tetracosyl ketone, butyl pentacosyl ketone, butyl hexacosyl ketone, butyl heptacosyl ketone, butyl octacosyl ketone, butyl nonacosyl ketone, butyl triacontyl ketone, etc.

A number of ketones containing at least 12 carbon atoms are availble as mixtures which are either products or by-products of commercial operations. These mixtures generally are available at comparatively low cost and, as another advantage of the present invention, the mixtures may be used without the added time and expense of separating specific compounds in pure state. One such mixture available commercially as a primary product of the process is stearone which is diheptadecyl ketone. Another such mixture is laurone which is dilauryl ketone.

As hereinbefore set forth in another embodiment of the invention the ketone may contain at least 8 carbon atoms. Illustrative ketones containing from 8 to 11 carbon atoms include methyl hexyl ketone, methyl heptyl ketone, methyl octyl ketone, methyl nonyl ketone, ethyl amyl ketone, ethyl hexyl ketone, ethyl heptyl ketone, ethyl oxtyl ketone, propyl butyl ketone, propyl amyl ketone, propyl hexyl ketone, propyl heptyl ketone, dibutyl ketone, butyl amyl ketone, butyl hexyl ketone, etc.

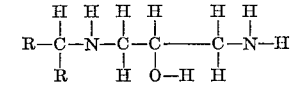
While the alkyl ketones are preferred, in some cases ketones containing unsaturation in the aliphatic group may be employed, but not necessarily with equivalent results. Furthermore, while it generally is preferred to utilize the same ketone in forming the di-substituted product, it is understood, in some cases, that different ketones may be employed, at least one and preferably both of the ketones containing at least 12 carbon atoms each. In this embodiment one ketone preferably is reacted first with 1,3-diamino-2-propanol and then the other ketone is reacted with the product of the first reaction, either in the same reaction zone or after removal of the first reaction product from the reaction zone. In some cases, the aliphatic ketone may contain a non-hydrocarbon substituent in the chain and this substituent may contain oxygen, nitrogen, sulfur, etc.

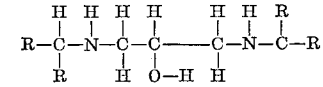
The reaction of 1,3-diamino-2-propanol and aliphatic ketone may be effected in any suitable manner. In one embodiment the diamino propanol and ketone are reacted by heating at refluxing conditions. This results in the preparation of a Schiff base, and the Schiff base may be utilized as the additive in the present invention. In another embodiment, the Schiff base is reduced, and this may be effected in any suitable manner by hydrogenating the Schiff base in the presence of hydrogen and a hydrogenation catalyst including nickel, platinum, palladium, etc., the hydrogenation generally being effected at a temperature of from about atmospheric to about 200° C., although higher temperatures may be employed in some cases. In still another embodiment, the reaction of 1,3-diamino-2-propanol and ketone may be effected in the presence of hydrogen and a hydrogenation catalyst in order to form the reductively alkylated product in one step.

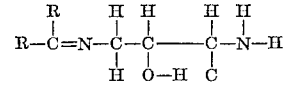
When desired, the reaction is effected in the presence of a solvent. Any suitable solvent may be employed and preferably comprises a hydrocarbon including benzene, toluene, xylene, ethyl benzene, cumene, decalin, naphtha, etc. The temperature of reaction will depend upon whether a solvent is employed and, when employed, upon the particular solvent. In general, the reaction is effected at a temperature of from about 80° to about 220° C. Water formed during the reaction may be removed in any suitable manner including, for example, by separating under reduced pressure, by removing an azeotrope of water-solvent, by distilling the reaction product at an elevated temperature, etc.

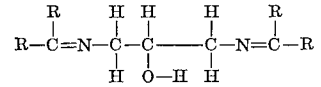
It will be noted that 1,3-diamino-2-propanol contains two primary amino groups (NH_2 groups) and that the reaction product may comprise the mono- and/or di-substituted diamino propanol. Generally, the di-substituted product is obtained, although the mono-substituted or a mixture of the mono- and di-substituted product may be prepared and utilized in the present invention. When the mono-substituted product is desired, one mol of ketone is reacted per mol of diamino propanol. When the di-substituted product is desired, two mols of ketone ara reacted per mol of the diamino propanol. In order to insure complete reaction, it is understod that an excess of the diamino alcohol or ketone may be supplied to the reaction zone, and the excess preferably is subsequently removed in any suitable manner.

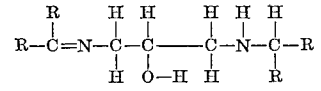
The composition of the reaction products of the present invention has not been completely established. It is believed that the reaction products may comprise compounds having one or more of the chemical structures illustrated by structures (1) through (5) below.


(1)


(2)

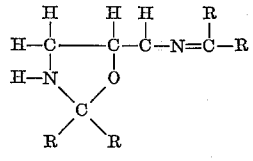

(3)


(4)

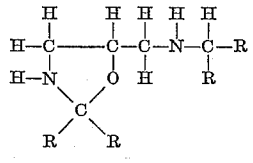

(5)

R in the above structures represents the residual aliphatic radicals derived from the ketone and will be the same or different composition and/or configuration depending upon the ketone used as the reactant. However, cyclization may occur during the reaction and the composition of the present invention may comprise com-

pounds of the general structure illustrated in structures (6) and (7).



(6)



(7)

Here again R represents the aliphatic radicals derived from the ketone.

It is understood that the composition of the present invention will comprise one of the compounds illustrated in the above structures or a mixture of two or more thereof.

From the above description, it will be noted that a number of different compounds may be prepared and utilized in accordance with the present invention. It is understood that, while all of these compounds are effective in certain substrates, they are not necessarily equivalent in the same or different substrate.

As hereinbefore set forth, the reaction products prepared in the above manner are new compositions of matter which possess unexpected properties over related but different compositions of matter of the prior art. Depending upon the reactants and conditions employed, the reaction product may comprise a specific compound, but generally will comprise a mixture of different compounds. Another advantage to the present invention is that the mixture of compounds prepared in the above manner may be utilized without the added expense and time of separating a specific compound from the mixture. The reaction products will range from liquids to solids and, when desired, may be prepared as solutions in suitable solvents for ease of handling and use.

The reaction product is recovered as a viscous liquid or solid. In some cases, the product will be marketed and utilized as a solution in a solvent. Conveniently, this solvent comprises the same solvent used in preparing the reaction product and is recovered in admixture with at least a portion of the solvent, thereby avoiding the necessity of removing all of the solvent and subsequently adding it back. When a more dilute solution is desired than is recovered in the manner hereinbefore set forth, it is understood that the same or different solvent may be co-mingled with the mixture to form a solution of the desired concentration.

The concentration of additive to be used in the organic substrate will depend upon the particular substrate and the particular benefits desired. In one embodiment the additive will be used in a concentration of from about 0.0001% to about 10% or more and more specifically in a concentration of from about 0.001% to about 5% by weight of the substrate. In most cases the additive will be used in a concentration of from about 0.001% to about 1% by weight of the substrate.

It is understood that the inhibitor of the present invention may be used along with other additives incorporated in the substrate including, for example, higher alcohols, esters, organic amines, polybutene, sulfurized fatty materials, sulfur-chlorine compounds, dyes, fillers, etc. In some cases it may be of advantage to also include a metal deactivator as, for example, disalicylal diaminopropane, etc., or to include other additives such as tricresyl phosphate, dialkylphenols, trialkyl phenols including 2,6-di-tert-butyl-4-methylphenol, 2,4-dimethyl-6-tert-butylphenol, dialkyl phenylene diamines, diarylphenylene diamines, diamino diphenyl methanes, tetraalkyl diamino-

diphenyl methanes, diphenylamine, alkylated diphenylamine, styrenated diphenylamine, aminophenols, alkylamino phenols, phenothiazine, organic selenium compounds, zinc dialkyldithiocarbamates, cadmium dialkyldithiocarbamates, naphthols, hydroxydiphenylamines, naphthyl-p-phenylene diamine, phenyl-alpha-naphthylamine, phenyl-beta-naphthylamine, alkoxy diphenylamine N-phenyl-N'-cyclohexyl-p-phenylene diamine, reaction products of sulfur dichloride or sulfur monochloride with alkylphenols, aniline, alkylanilines, reaction products of formaldehyde and alkylphenols, guaiacol, etc.

In other cases, especially in lubricating oils, the inhibitor of the present invention may be used in combination with one or more of bearing corrosion inhibitors, peroxide decomposers or antioxidants: zinc dialkyldithiophosphates, phosphorus pentasulfide-olefin reaction products, sulfur-olefin reaction products, sulfurized terpenes, aromatic hydroxysulfides and disulfides and their neutralization products with calcium, barium and magnesium oxides or hydroxides, calcium, barium or magnesium sulfonates, barium or calcium alkylphenates, etc., as well as additional corrosion inhibitors, extreme pressure additives, viscosity index improvers, detergents, etc. When desired, the inhibitor composition of the present invention may be prepared as a mixture with one or more of these other additives and incorporated in the substrate in this manner.

The additive may be incorporated in the substrate in any suitable manner. As hereinbefore set forth, the additive conveniently is marketed as a solution in a suitable solvent, including hydrocarbons and particularly aromatic hydrocarbons as benzene, toluene, xylene, cumene, etc. When the additive is to be incorporated in a liquid substrate, it may be added thereto in the desired amount and the resultant mixture suitably agitated in order to obtain intimate admixing of the additive in the substrate. When the additive is to be utilized as a corrosion inhibitor in plant equipment, it may be introduced into a fractionator, vapor line or at any other suitable point in order to prevent corrosion of the plant equipment. In this embodiment, the additive carries over into the product of the process and also serves therein as a beneficent. It is understood that a portion of the additive may be introduced into the plant equipment and an additional portion of the additive incorporated in the effluent product when so desired.

The following examples are introduced to illustrate further the novelty and utility of the present invention but not with the intention of unduly limiting the same.

EXAMPLE I

The reaction product of this example was prepared by the reaction of 1,3-diamino-2-propanol with methyl heptadecyl ketone. 45 g. of 1,3-diamino-2-propanol (0.5 mol+5 g. excess) was refluxed with 282 g. of methyl heptadecyl ketone (1 mol) in 200 g. toluene. 20.8 cc. of water was collected. The product then was reduced by hydrogenating at 120° C. in the presence of a platinum-containing catalyst. The basic mol combining weight found by titration with 0.1 N perchloric acid in glacial acetic acid-benzene was 327, the calculated for the disubstituted compound being 311. The product is a white powder, melting at 61–63° C. and has a specific gravity at 155° F. of 0.8584.

The reaction product prepared in the above manner was tested in a method referred to as "Erdco test." In this method, heated oil is passed through a filter, and the time required to develop a differential pressure across the filter of 25 in. Hg is determined. It is apparent that the longer the time, the more effective is the additive. However, with a very effective additive, the time to reach a differential pressure across the filter of 25 in. Hg is lengthened beyond reasonable limits so that the test is stopped after about 300 minutes and the differential pressure at that time is reported.

0.001% by weight of the reaction product prepared in

the above manner was incorporated in a commercial range oil and run in the "Erdco test." After 300 minutes the differential pressure across the filter was 0.8 in. Hg. On the other hand, a control sample (not containing this additive) developed a differential pressure across the filter of 25 in. Hg in 120 minutes. In still another test, 0.0005% of the same additive gave a differential pressure across the filter of 1.1 in. Hg after 300 minutes.

It will be noted that the additive of the present invention served to considerably retard deterioration of the range oil and thus will prevent clogging of burner tips, injectors, etc. during use of the oil.

EXAMPLE II

As hereinbefore set forth, the Schiff base may be utilized in the present invention. This Schiff base is formed in substantially the same manner as described in Example I, except that the final reducing with hydrogen is omitted. In the preparation used in the present example, 10 g. of 1,3-diamino-2-propanol (0.1 mol) was refluxed with 56.4 g. of methyl heptadecyl ketone (0.2 mol) in 100 g. toluene. 4.2 cc. of water was collected. Toluene was removed by heating under vacuum on a steam bath. The reaction product is a pale-tan powder, having a melting point of 54–56° C. and a specific gravity of 0.8763 at 155° F.

0.001% by weight of the reaction product prepared in the above manner was incorporated in another sample of the range oil described in Example I and run in the "Erdco test." After 300 minutes, the differential pressure across the filter was 0.8 in. Hg. Here again, it will be noted that the reaction product served to considerably retard deterioration of the range oil.

EXAMPLE III

Burner oils vary considerably, depending upon the geographic source of the burner oil and the treatment that the burner oil has received at the refinery. The burner oil used in Examples I and II developed 25 in. Hg in 120 minutes. In this example, a burner oil comprising a blend of 85% commercial fuel oil and 15% cracked gasoline was employed. When run in the "Erdco test" this oil developed a differential pressure of 25 in. Hg in about 50 minutes.

0.001% by weight of the reaction product prepared as described in Example II was incorporated in a sample of this blended burner oil and, when run in the Erdco Test, developed 0.4 in. Hg in 180 minutes. Because this burner oil is more difficult to stabilize, a period of 180 minutes is considered as satisfactory, and the test, therefore, was discontinued after such time. It will be noted that the reaction product was very effective in retarding deterioration of the blended burner oil.

EXAMPLE IV

Deterioration of burner oil also is evidenced by discoloration. In this example, a commercial fuel oil was stored for about 164 days at 100° F. and the color of the oil was determined after such storage. The color was determined in a Lumetron, Model 402–E, spectrophotometer. In this determination, the lower the number, the darker is the oil.

The additive used in this example was prepared by the reaction of 1,3-diamino-2-propanol with stearone, which is diheptadecyl ketone. 22.5 g. of 1,3-diamino-2-propanol (0.25 mol + 2.5 g. excess) was refluxed with 269 g. stearone (0.5 mol) in 200 g. of toluene. 10 cc. of water was collected. The product was reduced with hydrogen at 140° C. in the presence of a platinum-containing catalyst. The toluene solvent was evaporated by distilling under vacuum on a steam bath. The reaction product is a snow-white powder. Crystallized from Formula 30 alcohol, it melts at 72–75° C. The specific gravity at 155° F. is 0.8506.

0.01% of the reaction product prepared in the above manner was incorporated in a sample of a commercial fuel oil and, after storage at 100° F. for 165 days, the fuel oil had a color of 18. On the other hand, a control sample (not containing this additive) of the same oil after storage for 164 days at 100° F. had a color of 11. It will be noted that the reaction product of the present invention served to retard discoloration of the oil during storage.

EXAMPLE V

Another method of evaluating the additive is to store the same at 100° F. for a given time and determine the amount of sediment formed during such storage. In this example, 0.01% by weight of another portion of the additive prepared as described in Example II was incorporated in a sample of a commercial fuel oil. After 43 days in storage at 100° F., the oil had a sediment content of 0.8 mg. per 100 ml. On the other hand, a control sample of the same oil (not containing this additive) had a sediment content of 8.7 mg. per 100 ml. after storage at 100° F. for 43 days. Here again, it will be noted that the reaction product served to considerably improve the burner oil.

EXAMPLE VI

0.01% by weight of the additive prepared as described in Example I was incorporated in another sample of the fuel oil described in Example V. After 44 days storage at 100° F., the oil containing the additive had a sediment content of 0.5 mg. per 100 ml. This is a considerable reduction as compared to the 8.7 mg. per 100 ml. obtained with the control sample when stored at 100° F. for 43 days.

EXAMPLE VII

As hereinbefore set forth, the additive of the present invention also functions to retard corrosion. This was evaluated in a humidity cabinet test. In this test, a highly polished steel panel is dipped into a viscous naphthenic mineral oil, excess oil is drained, and the panel is placed in a humidity cabinet maintained at 120° F. in an atmosphere saturated with water. The panels are slowly rotated, and the days required for visible corrosion to appear on the panel is reported. A panel dipped in a control sample of the oil (not containing this additive) undergoes visible corrosion in 2–3 hours.

1% by weight of the additive prepared as described in Example I was incorporated in another sample of the oil. The panel dipped in this oil and then placed in the humidity cabinet did not undergo visible corrosion until after 9 days of exposure at 120° F. to the atmosphere saturated with water. Thus, it will be seen that the additive served to considerably reduce corrosion.

EXAMPLE VIII

As hereinbefore set forth, the reaction product of 1,3-diamino-2-propanol with an aliphatic ketone containing at least 8 carbon atoms possesses unexpectedly superior properties as compared with the reaction product of various other amino compounds with ketones. For example, an additive heretofore proposed for use in hydrocarbon distillates is prepared by the reaction of 2-hydroxy-acetophenone with 1,3-diamino-2-propanol. For comparative purposes this reaction product was evaluated in the Erdco method described above.

The reaction product of 1,3-diamino-2-propanol with 2-hydroxy-acetophenone was incorporated in a commercial JP–6 jet fuel in a concentration of 0.02% by weight. However, this additivce was not completely soluble at this concentration in the jet fuel. Accordingly, it was necessary to filter the jet fuel to remove sludge, water or extraneous impurities prior to the evaluation in the Erdco test.

The results of the evaluations in the Erdco test are reported in the following table. This table also reports the results of a run made in the same JP–6 jet fuel using the reaction product of Example I in order to compare these additives. In addition, the table reports the results of a blank or control sample (not containing an additive).

Table I

Additive	Differential Pressure In. Hg.	Minutes
None	25	31
0.02% by weight of the reaction product of Example I	0	300
0.02% by weight of the Schiff's base of 1,3-diamino-2-propanol with 2-hydroxy acetophenone	25	52

From the data in the above table it is seen that the reaction product of the present invention (reaction product of Example I) developed no pressure after 300 minutes. In contrast, the Schiff's base of 1,3-diamino-2-propanol with 2-hydroxy acetophenone developed a differential pressure of 25 in. Hg within 52 minutes.

The above data confirm the criticality of the inhibitor of the present invention in requiring the use of an aliphatic ketone in preparing the inhibitor in contrast to the cyclic ketone illustrated by acetophenone.

EXAMPLE IX

This example reports a comparison when using various ketones in the reaction with 1,3-diamino-2-propanol. The evaluations reported in the following table were made in the Erdco test hereinbefore described and in the same JP–6 jet fuel as used in Example VIII.

Table II

Additive	Differential Pressure In. Hg	Minutes
None	25	31
0.02% by weight of the reaction product of Example I	0	300
0.02% by weight of the reductive alkylation product of 1,3-diamino-2-propanol with ethylamyl ketone	21	300
0.02% by weight of the reductive alkylation product of 1,3-diamino-2-propanol with methyl ethyl ketone	25	32

The above data further demonstrate the requirement that the ketone used in the preparation of the inhibitor of the present invention must contain at least 8 and preferably at least 12 carbon atoms per molecule. It will be noted that the reaction product with methyl ethyl ketone (4 carbon atoms) developed a differential pressure of 25 in. Hg in 32 minutes. In contrast, the reaction product of Example I developed no differential pressure after 300 minutes.

The reaction product using ethyl amyl ketone (8 carbon atoms) developed a differential pressure of 21 in. Hg in 300 minutes. While this is considerably improved as compared to the results obtained with the reaction product using methyl ethyl ketone, it is not as good as the results obtained when using the reaction product prepared from methyl heptadecyl ketone (reaction product of Example I). Accordingly, for such use it is preferred that the ketone contains at least 12 carbon atoms.

EXAMPLE X

Another important criterion in evaluating additives for hydrocarbon distillates in the Erdco test is the rating of the preheater tube. This rating is determined according to a C.R.C. scale and reports the condition of the tube in each 1 inch increments. The total length of the tube is 13 inches. The tube ratings for some of the runs reported in Tables I and II above are shown in Table III below. The rating scale is as follows:

0—No visible deposit
1—Visible haze or dulling but no visible color
2—Discoloration barely visible
3—Light tan to peacock stain
4—Heavier than 3

Table III

Additive:	Preheater tube rating by inches
None	4444442000000
0.2% by weight of the reaction product of Example I	0000000000000
0.02% by weight of the Schiff's base of 1,3-diamino-2-propanol with 2-hydroxy-acetophenone	4444443200000
0.2% by weight of the reaction product of 1,3-daimino-2-propanol with methyl ethyl ketone	4444442000000

From the data in the above table it will be seen that all 13 inches of the tube were rated 0 (no visible deposit) for the run made with the reaction product of the present invention. In contrast, the control sample and the runs with the other reaction products all showed a rating of 4 for the first 6 inches of the tube. The tube cleanliness is an important evaluation because the build-up of deposits in the tube eventually will plug the tube and interefere with satisfactory operation of the equipment.

EXAMPLE XI

As hereinbefore set forth, the use of 1,3-diamino-2-propanol with ketones having at least 8 carbon atoms is unique and surprisingly superior to the reaction product of other amino compounds. The present example compares the results when using ethanol amine as a reactant instead of 1,3-diamino-2-propanol. Normally it would be expected that the reductively alkylated products of these alkanol amines would be substantially equivalent. However, as shown by the data in the following table, the results are considerably different.

These runs were made according to the Erdco method hereinbefore described and using a commercial range oil similar to but different from that used in Example I.

Table IV

Additive	Differential Pressure In. Hg	Minutes
None	25	65
0.001% by weight of the reductive alkylation product of 1,3-diamino-2-propanol with methyl heptadecyl ketone (reaction product of Example I)	0.8	300
0.001% by weight of the Schiff's base of 1,3-diamino-2-propanol with methyl heptadecyl ketone	0.8	300
0.001% by weight of the reductive alkylation product of 1,3-diamino-2-propanol with Stearone (reaction product of Example IV)	8.3	300
0.01% by weight of the reductive alkylation product of ethanol amine with methyl pentadecyl ketone	25	214
0.01% by weight of the reductive alkylation product of ethanol amine with Laurone (dilauryl ketone)	25	184
0.001% by weight of the reductive alkylation product of ethanol amine with Stearone	25	60

From the data in the above table it is seen that the use of ethanol amine is not equivalent to the use of 1,3-diamino propanol in the preparation of the inhibitor. It is to be noted that the reaction products prepared from 1,3-diamino-2-propanol developed very low differential pressures after 300 minutes when used in the low concentration of 0.001% by weight. In contrast, the reductive alkylation products prepared from ethanol amine developed 25 in. Hg differential pressure in less than 300 minutes, even when the additive was used in a concentration of 0.01% by weight which is 10 times greater than the 0.001% used with the inhibitors of the present invention.

EXAMPLE XII

This example compares the results obtained when using the reaction product of the present invention and when using the reductive alkylation product of tetraethylene

pentamine. Tetraethylene pentamine is representative of the alkylene polyamines heretofore proposed for reaction with ketones to prepare additives for organic substrates. The results are reported in the following table. Also included in the table is the result obtained when using the reductive alkylation product of ethanol amine.

The runs reported in the following table were made in the Erdco test described above when using a blended burner oil comprising 85% commercial fuel oil and 15% cracked gasoline, which blended oil is the same as used in the runs described in Example III of the present application.

Table V

Additive	Differential Pressure In. Hg	Minutes
None	25	50
0.001% by weight of the reductive alkylation product of tetraethylene pentamine with methyl pentadecyl ketone	25	156
0.001% by weight of the reductive alkylation product of tetraethylene pentamine with methyl heptadecyl ketone	25	165
0.001% by weight of the reductive alkylation product of ethanol amine with methyl pentadecyl ketone	25	41
0.001% by weight of the Schiff's base of 1,3-diamino-2-propanol with methyl heptadecyl ketone (reaction product of Example II)	0.4	180

As stated in Example III, this burner oil is more difficult to stabilize, and a period of 180 minutes is considered as satisfactory. Accordingly, the test was discontinued after such time.

From the data in the above table it will be noted that the reductive alkylation products using tetraethylene pentamine or ethanol amine all reached a differential pressure of 25 in. Hg in less than 180 minutes. In contrast, the reductive alkylation product of the present invention developed a differential pressure of only 0.4 in. Hg after 180 minutes.

I claim as my invention:

1. Hydrocarbon oil normally tending to deteriorate containing an inhibitor comprising the product resulting from the reaction of 1,3-diamino-2-propanol with an alkyl ketone having at least 8 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

2. Hydrocarbon oil normally tending to deteriorate containing an inhibitor comprising the product resulting from the reaction of 1,3-diamino-2-propanol with an alkyl ketone having at least 12 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

3. Hydrocarbon oil heavier than gasoline normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with an alkyl ketone having at least 8 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

4. Hydrocarbon oil heavier than gasoline normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with an alkyl ketone having at least 12 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

5. Burner oil normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with an alkyl ketone having at least 12 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

6. Jet fuel normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with an alkyl ketone having at least 12 carbon atoms in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

7. Burner oil normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with methyl heptadecyl ketone in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

8. Burner oil normally tending to deteriorate containing an inhibitor comprising the product resulting from the reductive alkylation of 1,3-diamino-2-propanol with diheptadecyl ketone in a proportion of from 1 to 2 mols of ketone per mol of diamino propanol.

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