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STABILIZATION OF BURNER OIL

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This invention relates to the stabilization of burner oils and more particularly to a novel method of retarding deterioration of burner oils in storage.

The term "burner oil" is used in the present specifications and claims to include hydrocarbon distillates boiling within the range of from about 300° F. to about 750° F. These burner oils are marketed under various trade names such as burner oil, fuel oil, furnace oil, diesel oil, etc., and are used principally in burner systems, diesel and combustion engines, and various other industrial and domestic equipment. Also specifically included in this classification are jet fuels or other hydrocarbon distillates containing components boiling within the range hereinbefore set forth and also containing lower boiling components. Jet fuels, for example, may have an initial boiling point as low as about 60° F. and an end boiling point within the range of from about 450° F. to about 600° F. or higher.

In storage the burner oil undergoes deterioration, with the formation of sediment, undesired discoloration, etc. The formation of sediment is objectionable because the sediment tends to plug strainers, burner tips, injectors, etc., and, when used as diesel fuel, tends to form varnish and sludge in the diesel engine. Discoloration of burner oils is objectionable for various reasons, including the customers' preference for light colored oils. Deterioration of jet fuel and burner oil at high temperature also is a serious problem. For example, jet fuel is used as a heat exchange medium for hot exhaust gases, and deterioration of the jet fuel results in plugging of the exchanger coils.

Burner oils present a difficult problem because of the wide variations in stability, apparently due to the many different sources from which burner oils are obtained. The formation of sediment and discoloration in storage vary considerably with the geographic source of the burner oil and with the treatment that the burner oil has received at the refinery. At present burner oils come principally from non-destructive distillation of petroleum oil and are commonly referred to in the art as straight run distillate, and from non-catalytic and catalytic cracking processes and are commonly referred to in the art as cycle stocks, the latter term being used because the burner oil is separated from a fraction which otherwise would be recycled to the cracking process for further conversion therein. Other sources of burner oils may include those produced by the reaction of carbon monoxide with hydrogen, in such processes as the Fischer-Tropsch process, synthesis process, Oxo process, etc. The stability problems of burner oils become complicated further when the burner oil comprises a blend of two or more different oils. The present invention is directed to a method of retarding deterioration of the burner oil by incorporating therein a particular type of additive.

While the improvement desired in burner oils may take the form of reduced sediment formation, retarded discoloration, etc., as hereinbefore set forth, satisfactory improvement also may be obtained in a somewhat different manner. One of the primary objectives is that the burner oil will not clog strainers, burner tips, injectors, etc., as hereinbefore set forth, and this objective also may be attained by suitably dispersing the particles in the burner oil so that they will be small enough to pass through said restricted zones without clogging thereof. Therefore, while the actual sediment content may be the same or larger, it is present in such a finely divided form

that the burner oil may be satisfactorily used without experiencing clogging difficulties. It has been found and will be shown in the following examples that the additives of the present invention effect improvements in certain fuel oils by this method.

In one embodiment the present invention relates to a method of retarding deterioration of burner oil which comprises incorporating therein a stabilizing concentration of the reaction product of an amine compound, having at least 12 carbon atoms and a straight chain of at least three carbon atoms attached to the nitrogen atom, with an epihalohydrin compound.

In a specific embodiment the present invention relates to a method of retarding deterioration of burner oil which comprises incorporating therein from about 0.0001% to about 1% by weight of the reaction product of tallow amine with epichlorohydrin.

In another specific embodiment the present invention relates to a method of retarding deterioration of jet fuel which comprises incorporating therein from about 0.0001% to about 1% by weight of the reaction product of di-(octadecyl) amine with epichlorohydrin.

In still another embodiment the present invention relates to a burner oil tending to deteriorate in storage containing, as an additive to retard said deterioration, a stabilizing concentration of the reaction product herein set forth.

It is essential in the present invention that the amine compound used in preparing the reaction product contains at least 12 carbon atoms and preferably at least 15 carbon atoms. Generally the total number of carbon atoms in the amine will not exceed about 40 carbon atoms per molecule. Furthermore, the amine must contain a straight chain of at least three carbon atoms attached to the nitrogen atom. As will be shown in the appended examples, reaction products prepared from amines containing less than 12 carbon atoms are not satisfactory for effecting the desired improvements in the burner oil and, therefore, are not suitable for use in the present invention. Furthermore, it is essential that a straight chain of at least 3 carbon atoms must be attached to the nitrogen atom of the amine in order to obtain the improved results of the present invention. In other words, the alkyl group attached to the nitrogen atom is of normal configuration and not secondary, tertiary, or of cyclic configuration. However, the alkyl group may contain branching in the chain provided such branching occurs on the fourth carbon atom from the nitrogen atom or further distant therefrom. As also will be shown in the appended examples, reaction products prepared from amines having branched configuration do not give satisfactory results and, therefore, are not suitable for use in the present invention.

Any suitable alkyl amine meeting the requirements set forth may be used in preparing the additive of the present invention. In addition to the above requirements, it is essential that the alkyl amine is a primary or secondary amine, that is, only one or two of the hydrogen atoms attached to the nitrogen atom are substituted by alkyl groups. Tertiary amines (no hydrogen atoms attached to the nitrogen atom) cannot be used in the present invention. It is understod that the term "alkyl amine" is used in the present specifications and claims to include primary alkyl amines, secondary alkyl amines, polyamines, N-alkyl polyamines, N,N'-dialkyl polyamines, etc., all of which meet the requirements hereinbefore set forth.

Illustrative examples of primary alkyl amines include dodecyl amine, tridecyl amine, tetradecyl amine, pentadecyl amine, hexadecyl amine, heptadecyl amine, octadecyl amine, nonadecyl amine, eicosyl amine, heneicosyl amine, docosyl amine, tricosyl amine, tetracosyl amine,

pentacosyl amine, hexacosyl amine, heptacosyl amine, octacosyl amine, nonacosyl amine, triacontyl amine, hentriacontyl amine, dotriacontyl amine, tritriacontyl amine, tetratriacontyl amine, pentatriacontyl amine, hexatriacontyl amine, heptatriacontyl amine, octatriacontyl amine, nonatriacontyl amine, tetracontyl amine, etc. Conveniently the long chain amines are prepared from fatty acids or more particularly mixtures of fatty acids formed as products or by-products. Such mixtures are available commercially, generally at lower prices and, as another advantage of the present invention, the mixtures may be used without the necessity of separating individual amines in pure state.

As an example of such a mixture is hydrogenated tallow amine which is available under various trade names including "Alamine H26D" and "Armeen HTD." These products comprise mixtures predominating in alkyl amines containing 16 to 18 carbon atoms per alkyl group, although they contain a small amount of alkyl groups having 14 carbon atoms, and also meet the other requirements hereinbefore set forth.

Illustrative examples of secondary amines include di(dodecyl) amine, di(tridecyl) amine, di(tetradecyl) amine, di(pentadecyl) amine, di(hexadecyl) amine, di-(heptadecyl) amine, di(octadecyl) amine, di(nonadecyl) amine, di(eicosyl) amine, etc. In these examples each of the alkyl substituents contains a straight chain of at least three carbon atoms attached to the nitrogen atom. In another embodiment, which is not necessarily equivalent, the secondary amine will contain one alkyl group having at least 12 carbon atoms and another chain having less than 12 carbon atoms, both of the alkyl groups having a straight chain of at least three carbon atoms attached to the nitrogen atom. Illustrative examples of such compounds include N - propyl - dodecyl amine, N - butyl - dodecyl amine, N - amyl - dodecyl amine, N - butyl - tridecyl amine, N - amyl - tridecyl amine, etc., N - propyl - tetradecyl amine, N - butyl - tetradecyl amine, N-amyl-tetradecyl amine., etc. Here again, mixtures of secondary amines are available commercially, usually at a lower price, and such mixtures may be used in accordance with the present invention, provided that the amines meet the requirements hereinbefore set forth. An example of such a mixture available commercially is "Armeen 2HT" which consists primarily of dioctadecyl amine and dihexadecyl amine.

Preferred examples of N-alkyl polyamines comprise N-alkyl-1,3-diaminopropanes in which the alkyl group contains at least 12 carbon atoms and a straight chain of at least three carbon atoms attached to the nitrogen atom. Illustrative examples include N-dodecyl-1,3-diaminopropane, N-tridecyl-1,3-diaminopropane, N-tetradecyl - 1,3 - diaminopropane, N - pentadecyl - 1,3 - diaminopropane, N - hexadecyl - 1,3 - diaminopropane, N - heptadecyl - 1,3 - diaminopropane, N - octadecyl-1,3 - diaminopropane, N - nonadecyl - 1,3 - diaminopropane, N - eicosyl - 1,3 - diaminopropane, N - heneicosyl - 1,3 - diaminopropane, N - docosyl - 1,3 - diaminopropane, N-tricosyl-1,3-diaminopropane, N-tetracosyl - 1,3 - diaminopropane, N - pentacosyl - 1,3 - diaminopropane, N - hexacosyl - 1,3 - diaminopropane, N - heptacosyl - 1,3 - diaminopropane, N - octacosyl-1,3 - diaminopropane, N - nonacosyl - 1,3 - diaminopropane, N - triacontyl - 1,3 - diaminopropane, N - hentriacontyl - 1,3 - diaminopropane, N - dotriacontyl - 1,3-diaminopropane, N - tritriacontyl - 1,3 - diaminopropane, N-tetratriacontyl-1,3-diaminopropane, N-pentatriacontyl - 1,3 - diaminopropane, N - hexatriacontyl - 1,3-diaminopropane, N - heptatriacontyl - 1,3 - diaminopropane, N-octatriacontyl-1,3-diaminopropane, N-nonatriacontyl - 1,3 - diaminopropane, N - tetracontyl - 1,3 - diaminopropane, etc. As before, mixtures are available commercially, usually at lower prices, of suitable compounds of this class and advantageously are used for the purposes of the present invention. One such mixture is "Duomeen T" which is N-tallow-1,3-diaminopropane and predominates in alkyl groups containing from 16 to 18 carbon atoms each, although the mixture contains a small amount of alkyl groups containing 14 carbon atoms each. Another mixture available commercially is N-coco-1,3-diaminopropane which contains alkyl groups predominating in 12 to 14 carbon atoms each. Still another example is N-soya-1,3-diaminopropane which predominates in alkyl groups containing 18 carbon atoms per group, although it contains a small amount of alkyl groups having 16 carbon atoms. However, such mixtures can be used only if they do not contain a branched chain configuration in proximity to the nitrogen atoms, as hereinbefore set forth.

While the N-alkyl-1,3-diaminopropanes are preferred compounds of this class, it is understood that suitable N-alkyl ethylene diamines, N-alkyl-1,3-diaminobutanes, N-alkyl-1,4-diaminobutanes, N-alkyl-1,3-diaminopentanes, N - alkyl - 1,4-diaminopentanes, N-alkyl-1,5-diaminopentanes, N - alkyl-1,3-diaminohexanes, N-alkyl-1,4-diaminohexanes, N - alkyl - 1,5 - diaminohexanes, N-alkyl-1,6-diaminohexanes, etc. may be employed, but not necessarily with equivalent results. Also, it is understood that polyamines containing three or more nitrogen atoms may be employed provided they meet the requirements hereinbefore set forth. Illustrative examples of such compounds includes N - dodecyl - diethylene triamine, N-tridecyl-diethylene triamine, N-tetradecyl-diethylene triamine, etc., N - dodecyl - dipropylene triamine, N-tridecyl-dipropylene triamine, N-tetradecyl-dipropylene triamine, etc., N-dodecyl-dibutylene triamine, N-tridecyl-dibutylene triamine, N-tetradecyl-dibutylene triamine, etc., N-dodecyl-triethylene tetramine, N-tridecyl-triethylene tetramine, N-tetradecyl triethylene tetramine, etc., N-dodecyl-tripropylene tetramine, N-tridecyl-tripropylene tetramine, N-tetradecyl-tripropylene tetramine, etc., N-dodecyl-tributylene tetramine, N-tridecyl-tributylene tetramine, N-tetradecyl-tributylene tetramine, etc., N-dodecyl-tetraethylene pentamine, N-tridecyl-tetraethylene pentamine, N-tetradecyl-tetraethylene pentamine, etc., N-dodecyl-tetrapropylene pentamine, N-tridecyl-tetrapropylene pentamine, N-tetradecyl-tetrapropylene pentamine, etc., N-dodecyl-tetrabutylene pentamine, N-tridecyl-tetrabutylene pentamine, N-tetradecyl-tetrabutylene pentamine, etc.

In another embodiment, polyaminoalkanes meeting the requirements hereinbefore set forth may be employed but generally such materials are not available commercially and, therefore, generally are not preferred. Illustrative examples of such compounds includes 1,12-diaminododecane, 1,13-diamino-tridecane, 1,14-diamino-tetradecane, etc.

In general, it is preferred that the amine compound is a saturated compound and does not contain double bonds in the chain. However, in some cases, unsaturated compounds may be employed, provided they meet the other requirements hereinbefore set forth, although not necessarily with equivalent results. Such amine compounds may be prepared from unsaturated fatty acids and, therefore, may be available commercially at lower cost. Illustrative examples of such amine compounds include dodecylenic amine, didodecylenic amine, N-dodecylenic ethylene diamine, N-dodecylenic-1,3-diaminopropane, oleic amine, dioleic amine, N-oleic ethylene diamine, N-oleic-1,3-diaminopropane, linoleic amine, dilinoleic amine, N-linoleic ethylene diamine, N-linoleic-1,3-diaminopropane, etc. It is understood that these amine compounds are included in the present specifications and claims by reference to amine or amine compounds.

As hereinbefore set forth, two different amines may be reacted with the epihalohydrin compound. At least one of the amines must meet the qualifications hereinbefore set forth. The other amine may comprise any suitable compound containing primary and/or secondary amino groups. Preferred compounds comprise ethylene diamine, diethylene triamine, triethylene tetramine, tetraethylene pentamine, etc., similar propylene and polypropylene poly-

amines, butylene and polybutylene polyamines, etc. In still another embodiment, other suitable nitrogen-containing compounds may be used as, for example, urea, monoethanol amine, etc.

As hereinbefore set forth, the amine compound is reacted with an epihalohydrin compound. Epichlorohydrin is preferred. Other epichlorohydrin compounds include 1,2-epi-4-chlorobutane, 2,3-epi-4-chlorobutane, 1,2-epi-5-chloropentane, 2,3-epi-5-chloropentane, etc. In general, the chloro derivatives are preferred, although it is understood that the corresponding bromo and iodo compounds may be employed. In some cases epidihalohydrin compounds may be utilized. It is understood that the different epihalohydrin compounds are not necessarily equivalent in the same or different substrate and that, as hereinbefore set forth, epichlorohydrin is preferred.

In general, one or two mols of amine compound is reacted with one or two mols of epihalohydrin compound. It is understood that, in some cases, an excess of amine or of epihalohydrin may be supplied to the reaction zone in order to insure complete reaction, the excess being removed subsequently in any suitable manner. When two mols of amine are reacted per mol of epihalohydrin compound, the amine may comprise the same or different amine compound.

The desired quantity of alkyl amine and epihalohydrin compounds may be supplied to the reaction zone and therein reacted, although generally it is preferred to supply one reactant to the reaction zone and then introduce the other reactant step-wise. Thus, usually it is preferred to supply the alkyl amine to the reaction zone and to add the epihalohydrin compound step-wise, with stirring. When it is desired to react two different alkyl amines with the epihalohydrin compound, one of the amines is supplied to the reaction zone, the epihalohydrin compound added gradually, and the reaction completed, followed by the addition of the second alkyl amine. Generally, it is preferred to utilize a solvent and, in the preferred embodiment, a solution of the alkyl amine in a solvent and a separate solution of the epihalohydrin compound in a solvent are prepared, and these solutions then are commingled in the manner hereinbefore set forth. Any suitable solvent may be employed, a particularly suitable solvent comprising an alcohol including ethanol, propanol, butanol, etc., 2-propanol being particularly desirable.

The reaction is effected at any suitable temperature, which generally will be within the range of from about 20° to about 100° C. and preferably is within the range of from about 50° to about 75° C. A higher temperature range of from about 30° to about 150° C. or more, and preferably of from about 50° to about 100° C., is specified when the reaction is effected at superatmospheric pressure to increase the reaction velocity. Conveniently, this reaction is effected by heating the alkyl amine solution in dilute alcohol at refluxing conditions, with stirring, gradually adding the epihalohydrin compound thereto, and continuing the heating until the reaction is completed.

Either before or after removal of the reaction product from the reaction zone, the product is treated to remove halogen, generally in the form of an inorganic halide salt as, for example, the hydrogen halide salt. This may be effected in any suitable manner and generally is accomplished by reacting the product with a strong inorganic base such as sodium hydroxide, potassium hydroxide, etc., to form the corresponding metal halide. The reaction to form the metal halide generally is effected under the same conditions as hereinbefore set forth. After this reaction is completed, the metal halide is removed in any suitable manner, including filtering, centrifugal separation, etc. It is understood that the reaction product also is heated sufficiently to remove alcohol and water and this may be effected either before or after the treatment to remove the inorganic halide.

The reaction products prepared in the above manner are new compositions of matter and possess unexpected properties over related but different compositions of matter of the prior art. Depending upon the reactants and conditions employed, the reaction product generally will comprise a mixture of different compounds, which mixture may include polymeric 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 a solution in a suitable solvent for ease of handling and using.

In still another embodiment, after the reaction product of an alkyl amine and epihalohydrin is prepared, the reaction product may be reacted with other nitrogen-containing compounds including, for example, alkanol amines, urea, etc., instead of with the same or different alikyl amine as hereinbefore described. Illustrative alkanol amines include ethanol amine, propanol amine, butanol amine, pentanol amine, hexanol amine, etc.

As hereinbefore set forth, the reaction product prepared in the above manner is particularly advantageous for use as an additive in burner oil. It is incorporated in the burner oil in a stabilizing concentration which generally will be below about 1% by weight and usually in a concentration within the range of from about 0.0001% to about 1% by weight. It is understood that this additive may be used in conjunction with other additives, such as metal deactivators, antioxidants, synergists, cetane improvers, rust inhibitors, etc. Furthermore, it is understood that the additive may be prepared as a solution in a suitable solvent. In some cases, one or more of the other additives to be incorporated in burner oil is prepared as a solution in the solvent and, when desired, the additive of the present invention may be prepared as a mixture with one or more other additives, preferably as a solution in a suitable solvent, and the same marketed as a single commodity of multiple purposes.

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 two mols of hydrogenated tallow amine (Alamine H26D) with one mol of epichlorohydrin. It will be noted that the tallow amine is a mixture of primary amines predominating in 16 to 18 carbon atoms per alkyl group. The amine was prepared as a dilute solution in 2-propanol and was supplied to an autoclave and heated to reflux, with stirring. One mol of epichlorohydrin, separately prepared as a solution in 2-propanol, was added gradually to the autoclave and the heating and mixing were continued for about 1.5 hours to insure complete reaction. Thereafter, one mol of sodium hydroxide was added to the autoclave, and the heating and stirring were continued. After completion of the reaction, the mixture was filtered hot to remove sodium chloride, and the filtrate then was distilled to remove the alcohol solvent. The remaining product was a cream colored solid and was recrystallized from hot ethanol to give a white amorphous solid having a softening point of 45° C. and melting at about 67° C. to a clear light yellow liquid, which is soluble in benzene, 2-propanol, etc.

The reaction product prepared in the above manner was tested in a method referred to as the "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 that the test is

stoppped 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 sample of commercial range oil and run in the "Erdco test." After 300 minutes, the differential pressure across the filter was 0.6 in. Hg. On the other hand, a control sample (not containing this additive) reached a differential pressure across the filter of 25 in. Hg in about 120 minutes.

EXAMPLE II

As hereinbefore set forth, it is essential that the alkyl amine contains at least 12 carbon atoms in the alkyl group and has a straight chain of at least 3 carbon atoms adjoining the nitrogen atom. This is illustrated by comparing the results of Example I with the results reported in this example of reaction products prepared from amines not meeting this requirement. All of the reactions were effected in substantially the same manner as described in Example I except, of course, for the amine utilized. They were prepared by the reaction of two mols of amine with one mol of epichlorohydrin. The first amine used is octyl amine which, it will be noted, does not contain at least 12 carbon atoms in the alkyl group. The second amine employed is a tertiary alkyl primary amine containing about 18 carbon atoms in each alkyl group and is available commercially as "Alkylamine JM." The third amine used is dodecyl aniline, in which, it will be noted, a nitrogen atom is attached to an aromatic configuration. The fourth amine is rosin amine, in which the nitrogen atom is attached to an alicyclic structure, this amine being available commercially under the trade name of "Rosin Amine D." Each additive was used in a concentration of 0.001% by weight of the range oil. The results of runs made in the "Erdco test" are reported in the following table:

Table I

No.	Amine used in preparation	Minutes/In. Hg
1	Octyl amine	135/25
2	Tertiary alkyl amine	128/25
3	Dodecyl aniline	118/25
4	Rosin amine	300/7.2

From the data in the above table, it will be noted that none of the above reaction products are satisfactory for use in preventing deterioration of range oil. In contrast, the use of the reaction product described in Example I resulted in only 0.6 in. Hg after 300 minutes.

EXAMPLE III

As hereinbefore set forth, effective additives are obtained by the reaction of epichlorohydrin with both an amine and another amine or other nitrogen-containing compound. In the preparations of the present example, one mol of hydrogenated tallow amine was reacted with one mol of epichlorohydrin and then the product was further reacted. In the first preparation, one mol of hydrogenated tallow amine was dissolved in 2-propanol and heated with stirring to 60°–65° C. One mol of epichlorohydrin dissolved in 2-propanol was added gradually to the amine solution. After the reaction was completed, the entire solution was removed from the reaction zone and was added gradually to a refluxing solution of one mol of monoethanolamine. Subsequently, one mol of sodium hydroxide was reacted with the mixture and, upon completion of the reaction, the mixture was filtered hot to remove sodium chloride and the filtrate then was allowed to cool. A precipitate separated out, was filtered off, and dried to remove adhering solvent. The product had a softening point of 41° C. and melts at 46–47° C. to pale yellow liquid, soluble in benzene, toluene, hot ethanol, hot 2-propanol, etc.

In the second preparation of the present example, the reaction product of hydrogenated tallow amine and epichlorohydrin was reacted further with one mol of urea, this reaction being effected in substantially the same manner as described in the previous paragraph. Upon cooling of the filtrate, a precipitate separated out, was filtered off, and dried to remove adhering solvent. The product had a softening point of 45° C. and melts at 51–52° C. to an opaque liquid, soluble in benzene, toluene, hot ethanol, hot 2-propanol, etc.

Two mols of a mixture of di-(octadecyl) amine and di-(hexadecyl) amine (Armeen 2HT) was reacted with one mol of epichlorohydrin and then the product was reached with 0.5 mol of ethylene diamine. The reaction was effected in substantially the same manner as described in the previous paragraphs. The final reaction product was a pale yellow soft, waxy solid, melting at 30–32° C. to pale yellow liquid which, upon heating to about 70° C. clears to a transparent yellow liquid. It is soluble in benzene, toluene, 2-propanol, hot ethanol, etc.

These additives each were used in another sample of the range oil described in Example I and were incorporated therein in a concentration of 0.001% by weight of the range oil. The results are reported in the following table:

Table II

No.	First amine	Second Nitrogen Compound	Min./In. Hg
5	Tallow amine	Monoethanol amine	300/0.4
6	do	Urea	300/0.3
7	Di(octadecyl)amine and di(hexadecyl)amine.	Ethylene diamine	300/0.3

From the data in the above table, it will be noted that effective additives are obtained by the further reaction of the first reaction product with a different nitrogen containing compound. It will be noted that in one case the second nitrogen containing compound is an alkanol amine; in another case, it is urea and, in the third case, it is a short chain alkylene polyamine. However, in all of these cases, the original amines employed contained an alkyl group of at least 12 carbon atoms.

EXAMPLE IV

This example illustrates the reaction of three mols of hydrogenated tallow amine with two mols of epichlorohydrin. This reaction was effected by adding gradually one mol of tallow amine in propanol solution to a stirred solution of two mols of epichlorohydrin in 2-propanol at 60°–65° C. After the addition of the primary amine, the mixture was refluxed and then the one mol of tallow amine was added gradually to the mixture, following which two mols of sodium hydroxide were added, and the mixture reacted and then filtered, followed by distillation to remove the solvent. This reaction product was a pale yellow solid, having a softening point of 55° C. and melting to a pale yellow opaque liquid at about 65° C. which, upon heating to 74° C., clears to a transparent yellow liquid, soluble in toluene, warm benzene, 2-propanol, hot ethanol, etc.

0.001% by weight of the reaction product described above was incorporated in another sample of the range oil. When run in the Erdco test, the differential pressure across the filter after 300 minutes was 0.3 in. Hg.

EXAMPLE V

As hereinbefore set forth, stability problems of different burner oils vary considerably. In the present 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. It will be noted that this blended burner oil is of considerably poorer stability than the range oil used in the previous examples.

A number of different reaction products were prepared and tested in this burner oil, and the results are reported

in the following table. These reaction products were prepared in substantially the same manner as described in the previous examples. In the interest of simplicity, the specific details of preparation are omitted, but the reactants and quantities thereof are shown in the table. The reaction products each were utilized in the fuel oil in a concentration of 0.001% by weight. These runs were discontinued after 180 minutes.

Table III

No.	First Amine		Second N-Containing Compound		Epichlorohydrin, Mols	Min./In. Hg
	Mols	Compound	Mols	Compound		
8	2	Tallow Amine		None	1	180/0.6
9	1	do	1	Urea	1	180/0.4
10	1	do	0.5	Tetraethylene pentamine.	1	180/0.7
11	1	do	1	do	1	180/0.9
12	3	do		None	2	180/0.4
13	2	N-tallow-1,3-diaminopropane.		do	2	180/0.3
14	1	Tallow Amine	1	Ethylene diamine	2	180/0.5
15	2	Di(octadecyl) amine and di-(hexadecyl) amine.	0.5	do	1	180/0.3

From the data in the above table, it will be noted that the various reaction products prepared in accordance with the present invention were effective in retarding deterioration of the burner oil.

EXAMPLE VI

In another method of evaluating the additives, samples of the burner oil were stored at 100° F. for about 45 days and the mg./100 ml. of sediment was determined after such storage. Representative additives prepared in the manner previously described were each utilized in a No. 2 commercial fuel oil. The additive was used in a concentration of 0.01% by weight of the fuel oil. The results are shown in the following table:

Table IV

No.	First Amine		Second Amine		Epichlorohydrin, Mols	Days Stored	mg./100 ml.
	Mols	Cmpd.	Mols	Cmpd.			
16		None		None	None	43	8.7
17	2	N-tallow-1,3-diamino propane.		do	2	46	0.5
18	1	Tallow Amine	0.5	Tetraethylene pentamine.	1	46	0.3
19	1	do	1	do	1	45	0.5

From the data in the above table, it will be note that a considerable reduction in the sediment formation was obtained by using the reaction products of the present invention.

Furthermore, when evaluated in a test wherein the different samples of the oil with and without additives, after storage at 100° F. for about 45 days, are passed through a 400 wire mesh screen, and the times for the successive 400 ml. portions to pass through the screen are recorded, it was found that the oil containing the additives of the present invention had times of about one-half or less than the sample of oil not containing the additives. This further shows that the additives serve to prevent plugging of filters, tips, ejectors, etc.

I claim as my invention:

1. Burner oil tending to deteriorate in storage having incorporated therein a stabilizing concentration of the reaction product, formed at a temperature of from about 20° to about 150° C., of from one to two mol proportions of an aliphatic amine compound having at least one hydrogen atom attached to a nitrogen atom, said amine compound consisting of carbon, hydrogen and nitrogen, and having at least twelve and up to about forty carbon atoms and a straight chain of at least three carbon atoms attached to the nitrogen atom, with one mol proportion of an epihalohydrin compound selected from the group consisting of epichlorohydrin, 1,2-epi-4-chlorobutane, 2,3-epi-4-chlorobutane, 1,2-epi-5-chloropentane, 2,3-epi-5-chloropentane and corresponding bromo and iodo compounds, and reacting with an inorganic base to remove halogen.

2. Burner oil tending to deteriorate in storage having incorporated therein a stabilizing concentration of the reaction product, formed at a temperature of from about 20° to about 150° C., of from one to two mol proportions of tallow amine with one mol proportion of epichlorohydrin, and reacting with sodium hydroxide to remove chloride.

3. Burner oil tending to deteriorate in storage having incorporated therein a stabilizing concentration of the reaction product, formed at a temperature of from about 20° to about 150° C., of from one to two mol proportions of di-(hexadecyl) amine with one mol proportion of epichlorohydrin, and reacting with sodium hydroxide to remove chloride.

4. Burner oil tending to deteriorate in storage having incorporated therein a stabilizing concentration of the reaction product, formed at a temperature of from about 200° to about 150° C., of from one to two mol proportions of di-(octadecyl) amine with one mol proportion of epichlorohydrin, and reacting with sodium hydroxide to remove chloride.

5. Burner oil tending to deteriorate in storage having incorporated therein a stabilizing concentration of the reaction product, formed at a temperature of from about 20° to about 150° C., of one mol of tallow amine with one mol of epichlorohydrin which is further reacted with one mol of a nitrogen-containing compound, selected from the group consisting of alkyl amines, diaminoalkanes, alkanolamines and urea, and reacting with an inorganic base to remove chloride.

6. The burner oil composition of claim 5 further characterized in that said nitrogen-containing compound is monoethanolamine.

7. The burner oil composition of claim 5 further characterized in that said nitrogen-containing compound is urea.

8. The burner oil composition of claim 5 further char-

acterized in that said nitrogen-containing compound is tetraethylene pentamine.

9. The burner oil composition of claim 5 further characterized in that said nitrogen-containing compound is ethylene diamine.

10. Hydrocarbon jet fuel tending to deteriorate in storage containing a stabilizing concentration of the reaction product, formed at a temperature of from about 20° to about 150° C., of from one to two mol proportions of an aliphatic amine compound having at least one hydrogen atom attached to a nitrogen atom, said amine compound consisting of carbon, hydrogen and nitrogen, and having at least twelve and up to about forty carbon atoms and a straight chain of at least three carbon atoms attached to the nitrogen atom, with one mol proportion of an epihalohydrin compound selected from the group consisting of epichlorohydrin, 1,2-epi-4-chlorobutane, 2,3-epi-4-chlorobutane, 1,2-epi-5-chloropentane, 2,3-epi-5-chloropentane and corresponding bromo and iodo compounds, and reacting with an inorganic base to remove halogen.

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