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SYNERGISTIC ANTI-ICING COMPOSITION

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ABSTRACT OF THE DISCLOSURE

Synergistic anti-icing composition of (1) alkyl acid phosphate salt of N-alkyl-diamino-alkane and alkyl acid phosphate salt of alkyl-monoamine and (2) polyhydroxy alcohol.

BACKGROUND OF THE INVENTION

A serious problem in the operation of automobiles is stalling of the engine due to the formation of ice in the carburetor throttle body and on the throttle plate. As is well known, at temperatures ranging from about 30° to about 60° F. at periods of relatively high humidities, such stalling has been encountered under idling or low load conditions. This is caused by the air-borne moisture undergoing freezing due to the refrigerating effect encountered in normal fuel vaporization within the carburetor. The ice formed on the throttle plate and adjacent carburetor walls restricts the narrow air openings and causes engine stalling.

The icing problem is of increasing importance because of the design of newer automobiles. For example, present cars do not have a manual throttle and therefore the operator of the car is no longer able to increase the idle speed during the warm up period to prevent such stalling. Furthermore, the increasing use of automatic transmissions adds to this problem because the idle speed must be kept low to avoid creeping and, accordingly, the idle speed is not sufficiently fast to avoid stalling due to icing. Still another development which appears to add to this problem is the increased volatility of commercial gasolines, because more frequent stalling is encountered with the more volatile fuels.

Various methods have been proposed to eliminate the stalling of automobile engines, including the use of additives. In one method, an alcohol is used but this has the objection of requiring large concentrations of the alcohol in order to obtain reasonably satisfactory anti-icing. In another method, various phosphate salts of amines have been proposed.

While the use of a specific mixture of certain phosphate-amine salts has proven to be quite effective in preventing stalling, it now has been found that the use of such mixture in conjunction with a polyhydroxy alcohol even further improves the anti-icing effectiveness in avoiding stalling.

DESCRIPTION OF THE INVENTION

In one embodiment the present invention relates to a synergistic anti-icing composition of (1) phosphate salt of N-alkyl-diamino-alkane and phosphate salt of alkyl-monoamine and (2) polyhydroxy alcohol.

In another embodiment the present invention relates to gasoline containing a stabilizing concentration of the synergistic composition herein set forth.

As hereinbefore set forth, the use of a specific mixture of certain phosphate-amine salts has proved quite effective in preventing stalling. However, it is desirable to even further improve the effectiveness of the mixture and this is accomplished in accordance with the present inven-

tion by the use of a polyhydroxy alcohol in conjunction with the mixture. In addition to the improved anti-icing properties, the composition of the present invention is very effective in inhibiting deposits on the walls of the carburetor.

As hereinbefore set forth, one component of the additive composition of the present invention is a blend of a polyhydroxy alcohol with a mixture of a phosphate salt of an N-alkyl-diamino-alkane and a phosphate salt of an alkyl-monoamine. In a preferred embodiment of an alkyl acid phosphate, which may be the same or different, is used in preparing the diamine salt and the monoamine salt. The alkyl acid phosphate includes both the alkyl acid orthophosphates and the alkyl acid pyrophosphates. In the alkyl acid orthophosphates, the mono-alkyl ester, dialkyl ester or a mixture thereof may be employed. In the alkyl acid pyrophosphates, the monoalkyl ester, dialkyl ester, trialkyl ester or mixtures thereof may be employed, the dialkyl ester being preferred, and the ester groups may be attached to the same or different phosphorus atom. Generally, however, this compound will be symmetrical and, thus, the alkyl ester groups will be attached to different phosphorus atoms.

Illustrative examples of preferred alkyl acid orthophosphates and pyrophosphates are set forth below. In general, it is preferred that alkyl moiety contains from about 3 to about 20 and more particularly from about 4 to about 15 carbon atoms each. Accordingly, particularly preferred alkyl acid orthophosphates include monobutyl acid orthophosphate, dibutyl acid orthophosphate, mixture of mono- and dibutyl acid orthophosphates, monopentyl acid orthophosphate, dipentyl acid orthophosphate, mixture of mono- and dipentyl acid orthophosphates, monohexyl acid orthophosphate, dihexyl acid orthophosphate, mixture of mono- and dihexyl acid orthophosphates, monoheptyl acid orthophosphate, diheptyl acid orthophosphate, mixture of mono- and diheptyl acid orthophosphates, monooctyl acid orthophosphate, dioctyl acid orthophosphate, mixture of mono- and dioctyl acid orthophosphates, monononyl acid orthophosphate, dinonyl acid orthophosphate, mixture of mono- and dinonyl acid orthophosphates, monodecyl acid orthophosphate, didecyl acid orthophosphate, mixture of mono- and didecyl acid orthophosphates, monoundecyl acid orthophosphate, diundecyl acid orthophosphate, mixture of mono- and diundecyl acid orthophosphates, monododecyl acid orthophosphate, didodecyl acid orthophosphate, mixture of mono- and didodecyl acid orthophosphates, monotridecyl acid orthophosphate, ditridecyl acid orthophosphate, mixture of mono- and ditridecyl acid orthophosphates, monotetradecyl acid orthophosphate, ditetradecyl acid orthophosphate, mixture of mono- and ditetradecyl acid orthophosphates, monopentadecyl acid orthophosphate, dipentadecyl acid orthophosphate, etc. It is understood that the alkyl moiety may be of straight or branched chain and that it may be of primary, secondary or tertiary configuration.

Preferred alkyl acid pyrophosphates include monobutyl acid pyrophosphate, dibutyl acid pyrophosphate, mixture of mono- and dibutyl acid pyrophosphates, monopentyl acid pyrophosphate, dipentyl acid pyrophosphate, mixture of mono- and dipentyl acid pyrophosphates, monohexyl acid pyrophosphate, dihexyl acid pyrophosphate, mixture of mono- and dihexyl acid pyrophosphates, monoheptyl acid pyrophosphate, diheptyl acid pyrophosphate, mixture of mono- and diheptyl acid pyrophosphates, monooctyl acid pyrophosphate, dioctyl acid pyrophosphate, mixture of mono- and dioctyl acid pyrophosphates, monononyl acid pyrophosphate, dinonyl acid pyrophosphate, mixture of mono- and dinonyl acid pyrophosphates, monodecyl acid pyrophosphate, didecyl acid pyrophosphate, mixture of mono- and didecyl acid pyrophosphates,

monoundecyl acid pyrophosphate, diundecyl acid pyrophosphate, mixture of mono- and diundecyl acid pyrophosphates, monododecyl acid pyrophosphate, didodecyl acid pyrophosphate, mixture of mono- and didodecyl acid pyrophosphates, monotridecyl acid pyrophosphate, ditridecyl acid pyrophosphate, mixture of mono- and ditridecyl acid pyrophosphates, monotetradecyl acid pyrophosphate, ditetradecyl acid pyrophosphate, mixture of mono- and ditetradecyl acid pyrophosphates, monopentadecyl acid pyrophosphate, dipentadecyl acid pyrophosphate, mixture of mono- and dipentadecyl acid pyrophosphates, etc. Here again, it is understood that the alkyl moiety may be of straight or branched chain and may be of primary, secondary or tertiary configuration.

The specific phosphates hereinbefore set forth are preferred. It is understood that other suitable alkyl phosphates may be used in accordance with the present invention. For example, alkyl acid phosphates, including both the ortho- and pyrophosphates, are manufactured commercially as a mixture of mono- and dialkyl acid phosphates and are available commercially at a considerably lower cost. In many cases, such mixtures are very suitable for use in preparing the salts of the present invention and such use, therefore, is preferred for economic reasons.

While the alkyl acid phosphates are preferred, it is understood that other suitable phosphates may be employed but not necessarily with equivalent results. For example, in place of the alkyl moiety of the phosphate, an unsaturated aliphatic group may be employed, and thus may contain a double bond in the aliphatic chain. In still another embodiment, the alkyl moiety or moieties may be replaced by cyclic derivatives including particularly cyclohexyl, but may comprise cyclobutyl, cyclopentyl, cycloheptyl, cyclooctyl, etc. It is understood that the aliphatic or cyclic groups may contain hydrocarbyl or non-hydrocarbyl substituents attached thereto, the last mentioned being selected from hydroxy, alkoxy, etc.

In another embodiment, the phosphate used in preparing the salt is an oxyalkylenated hydroxyhydrocarbon phosphate, including the mono- and di-(oxyalkylenated hydroxyhydrocarbon) phosphate and mixtures thereof. The number of oxyalkylene groups will range from 1 to 40 and preferably from 1 to 10 and may contain from 1 to 10 and preferably from 2 to 4 carbon atoms per oxyalkylene group. In one embodiment, the hydroxyhydrocarbon moiety used in preparing the oxyalkylenated hydroxyhydrocarbon phosphate is a mono- or polyalkylphenol in which each alkyl group contains from 1 to 30 carbon atoms. Illustrative preferred phosphates in this embodiment include mono- and/or di-(polyoxypropylenated hexylphenol) phosphate containing an average of about 12 oxyethylene groups, mono- and/or di-(polyoxypropylenated heptylphenol) phosphate containing an average of about 9 oxyethylene groups, mono- and/or di-(polyoxyethylenated octylphenol) phosphate containing an average of about 5 oxyethylene groups, mono- and/or di-(polyoxyethylenated nonylphenol) phosphate containing an average of about 2 oxyethylene groups, mono- and/or di-(monooxyethylenated decylphenol) phosphate, etc. It is understood that a mixture of these phosphates may be used.

In another embodiment, the oxyalkylenated hydroxyhydrocarbon phosphate is oxyalkylenated alkanol phosphate in which the alkanol moiety contains from 6 to 40 and preferably from 10 to 30 carbon atoms. The oxyalkylene groups will be the same as hereinbefore set forth. Here again, the mono- and/or di-(oxyalkylenated alkanol) phosphate may be used. Illustrative examples in this embodiment include the mono- and/or di-(polyoxypropylenated hexanol) phosphate containing an average of about 6 oxyethylene groups, mono- and/or di-(polyoxypropylenated octanol) phosphate containing an average of about 5 oxyethylene groups, mono- and/or di-(polyoxyethylenated decanol) phosphate containing an average of about 4 oxyethylene groups, mono- and/or di-

(polyoxyethylenated dodecanol) phosphate containing an average of about 3 oxyethylene groups, mono- and/or di-(polyoxyethylenated tetradecanol) phosphate containing an average of about 2 oxyethylene groups, mono- and/or di-(mono-oxyethylenated pentadecanol) phosphate, etc. It is understood that a mixture of these phosphates may be employed.

Any suitable alkyl monoamine is used in preparing the phosphate salt of alkyl monoamine for use as one component of the additive composition of the present invention. The alkyl monoamine may contain from about 3 to about 20 carbon atoms and preferably contains from about 4 to about 12 carbon atoms and thus includes butyl amine, pentyl amine, hexyl amine, heptyl amine, octyl amine, nonyl amine, decyl amine, undecyl amine, dodecyl amine, etc. It is understood that the alkyl moiety may be of straight or branched chain and may be of primary, secondary or tertiary configuration. A particularly preferred alkyl monoamine for use in the present invention is 2-ethylhexyl amine. Other preferred alkyl monoamines include those known as beta amines in which the alkyl group is attached to the nitrogen atom through the beta carbon of the alkyl group.

Any suitable diamine is used in preparing the phosphate salt for use as a component in the additive composition of the present invention. While the diamine may contain from about 3 to about 40 carbon atoms, it preferably contains from about 8 to about 20 carbon atoms. A particularly preferred diamine is N-alkyl diaminoalkane in which the alkyl moiety contains from about 3 to about 30 carbon atoms and more particularly from about 6 to about 20 carbon atoms and the alkane moiety contains from about 2 to about 12 carbon atoms and preferably from about 3 to about 6 carbon atoms. A particularly preferred N-alkyl diaminoalkane is N-alkyl-1,3-diaminopropane, the alkyl group being derived from tallow. This compound is available commercially under the trade name of "Duomeen T." Other preferred N-alkyl-1,3-diaminopropanes comprise those in which the alkyl group is derived from lauric acid, coconut fatty acid, soya fatty acid, etc. These are available commercially at the present time and comprise mixed alkyl-substituted 1,3-diaminopropanes. For example, in the case of "Duomeen T" the alkyl group contains from about 12 to 20 carbon atoms per group and mostly contain 16 to 18 carbon atoms. However, when desired, the alkyl group of the N-alkyl-1,3-diaminopropanes or other N-alkyl-diaminoalkanes may be prepared to contain any number of carbon atoms desired in the alkyl group and, thus, is selected from hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, nonadecyl, eicosyl, etc. It is understood that a mixture of diamines containing different alkyl groups may be employed and that the alkyl and alkane moieties may be of straight or branched chain. Furthermore, it is understood that the alkyl moiety may be of primary, secondary or tertiary configuration. Also, particularly preferred in this embodiment are the beta amines in which the alkyl group is attached to the nitrogen atom through the beta carbon atom of the alkyl group.

While the N-alkyl-1,3-diaminopropanes are preferred, it is understood that other suitable N-alkyl diaminoalkanes may be employed. Illustrative examples include N-alkyl-1,2-diaminoethanes, N-alkyl-1,2-diaminopropanes, N-alkyl-1,2-diaminobutanes, N-alkyl-1,3-diaminobutanes, N-alkyl-1,4-diaminobutanes, N-alkyl-1,2-diaminopentanes, N-alkyl-1,3-diaminopentanes, N-alkyl-1,4-diaminopentanes, N-alkyl-1,5-diaminopentanes, N-alkyl-1,2-diaminohexanes, N-alkyl-1,3-diaminohexanes, N-alkyl-1,4-diaminohexanes, N-alkyl-1,5-diaminohexanes, N-alkyl-1,6-diaminohexanes, etc.

In another embodiment the alkyl-monoamine and/or alkyl-diaminoalkane may contain a double bond in the alkyl group. In still another embodiment the alkyl group may contain non-hydrocarbyl substitutions, the substitu-

tions being selected from hydroxy, alkoxy, N-dihydrocarbylamide, halogen, particularly chlorine and fluorine, etc. In still another embodiment, in place of an aliphatic group, the amine may be a cyclic amine as, for example, cyclohexyl amine, dicyclohexyl amine and cyclohexyl-

diaminoalkane or the cycloalkyl group may be cyclobutyl, cyclopentyl, cycloheptyl, cyclooctyl, etc. In still another embodiment the cyclic group may be a heterocyclic nitrogen compound including piperidyl, piperazyl, etc. It is understood that the different amines are not necessarily equivalent.

In general, the neutral salts of the phosphate and alkyl monoamine and of the phosphate and diamine are preferred. The neutral salts are prepared by utilizing stoichiometric amounts of the acid and the amine. In other words, the concentration of the alkyl acid phosphate and of the alkyl monoamine or of the alkyl acid phosphate and of the diaminoalkane will be selected so that there will be an equivalent number of acid groups to amino groups. Thus, the specific concentrations will depend upon whether the orthophosphate or pyrophosphate or whether the monoalkyl phosphate or dialkyl phosphate are used. It also will depend upon whether the monoamine or diamine is employed. In another embodiment, the salt is a basic salt which is prepared by utilizing a deficiency of acid groups in relation to the amino groups. In still another embodiment, the acid salt is used, which is prepared by using an excess of acid groups with relation to the amine groups. From the above description it will be seen that a number of different salts may be used in accordance with the present invention. It is understood that these different salts are not necessarily equivalent in the same or different fuels but all of them will serve to give improved results in most, if not all, fuels.

The salts are prepared in any suitable manner and, in general, are prepared by mixing the phosphate and the alkyl monoamine and/or diaminoalkane in the appropriate proportions at ambient temperature, preferably with vigorous stirring. When desired, the phosphate salt of the monoamine and the phosphate salt of the diamine may be separately prepared and utilized as such or subsequently mixed to prepare a blended composition. In another embodiment the mixture of salts may be prepared simultaneously by reacting the alkyl phosphate or phosphates with both the monoamine and diamine in the required proportions to form the mixture of salts in one step.

The salts are readily prepared at room temperature, although slightly elevated temperature which generally will not exceed about 200° F. may be employed when desired. Excessive temperatures should be avoided in order to prevent the undesired formation of reaction products resulting in the liberation of water and in the formation of phosphor amidic acid derivatives or other undesired reaction products. When desired, it may be of advantage to utilize a solvent, either in forming a more fluid mixture of the phosphates and/or amines before mixing or during the mixing thereof. In some cases it is desirable to admix the salt or salts with a solvent in order to form a more fluid final product. Any suitable solvent may be used and generally will comprise an organic compound and more particularly a hydrocarbon distillate. Particularly preferred solvents are aromatic hydrocarbons including benzene, toluene, xylene, ethylbenzene, cumene, etc., or mixtures thereof, or paraffinic hydrocarbons including pentane, hexane, heptane, octane, nonane, decane, etc., or mixtures thereof, or mixtures of the aromatic and paraffinic hydrocarbons, including naphtha, gasoline, etc.

As hereinbefore set forth, the phosphate salt of monoamine and the phosphate salt of diamine may be prepared separately or in admixture. When prepared separately, the salts may be blended or the salts may be added separately to the substrate. The different salts will be used in any suitable proportion, which may range from 5% to 95% of one salt and 5% to 95% by weight of the other salt. In some cases it is preferred that the phosphate salt of

the diamine is used in a larger proportion and, in such cases, the salt of the diamine is used in a concentration of from about 60% to about 90% by weight and the phosphate salt of the monoamine is used in a concentration of from about 10% to about 40% by weight, exclusive of solvent. When a solvent is employed, a stock solution may be prepared to contain the active ingredients in a concentration of from about 10% to a saturated solution which will be above about 50% by weight of active ingredients.

The phosphate salts, whether prepared separately or simultaneously, are blended with a polyhydroxy alcohol. A preferred polyhydroxy alcohol is the commercially available hexylene glycol which is 2,4-dihydroxy-2-methylpentane. Other polyhydroxy alcohols include ethylene glycol, propylene glycol, butylene glycol, pentylene glycol, other hexylene glycols as, for example, 1,6-dihydroxyhexane, 2,6-dihydroxyhexane, etc., heptylene glycol, octylene glycol, etc., diethylene glycol, dipropylene glycol, dibutylene glycol, etc., dihydroxycyclohexane as, for example, 1,4-dihydroxycyclohexane, 1,3-dihydroxycyclohexane, 1,2-dihydroxycyclohexane, etc., glycerol, 1,2,3-butanetriol, erythritol, etc. In a preferred embodiment the polyhydroxy alcohol is a dihydroxy alcohol containing from 2 to 8 carbon atoms and may be of straight or branched chain. However, it is understood that the polyhydroxy alcohol may contain 3 or 4 hydroxy groups, as well as being of aliphatic or cyclic configuration.

The trihydroxy or tetrahydroxy alcohols are of limited solubility in the phosphate-amine salts, the degree of solubility varying with the particular phosphate-amine salts and the particular solvent used in preparing solutions of the composition. Accordingly, these polyhydroxy alcohols will be selected with reference to the particular phosphate-amine salts and solvents.

In another embodiment, a mixture of polyhydroxy alcohols is employed. Illustrative mixtures include a mixture comprising from 85% to 95% by weight of hexylene glycol and 5% to 10% by weight of glycerol, a mixture of 75% to 95% by weight of ethylene glycol and 5% to 25% by weight of erythritol, etc.

The mixture of phosphate-amine salts and polyhydroxy alcohol will be used in any suitable proportion, which may range from 5% to 95% and preferably from 10% to 90% by weight of the mixed phosphate-amine salts and from 95% to 5% and preferably from 90% to 10% by weight of the polyhydroxy alcohol.

The amount of total additive composition to be added to the gasoline will be sufficient to effect improved deicing. For economic reasons, the concentration should be as low as practicable and may range from 0.0001% to 0.05% by weight and preferably within the range of from about 0.002% to about 0.01% by weight of the fuel, based on the mixed phosphate-amine salts and polyhydroxy alcohol exclusive of solvent when employed. While each of the phosphate-amine salts and the polyhydroxy alcohol may be added separately to the fuel, or the mixed phosphate-amine salts and the polyhydroxy alcohol added separately to the fuel, it generally is preferred to prepare a composition of the phosphate-amine salts and polyhydroxy alcohol in the proper concentrations and to add this composition to the fuel in the desired amount. When desired, the mixture of amine salts and polyhydroxy alcohol may be prepared as a solution in a suitable solvent such as a paraffinic, aromatic and/or naphthenic naphtha or gasoline. When desired, the solvent may comprise an aromatic or paraffinic hydrocarbon and will be selected from those hereinbefore set forth. In such a solution, the phosphate-amine salts and hexylene glycol will comprise from about 10% to about 90% and preferably from about 25% to about 75% of the solution.

The novel additive mixture of the present invention also serves as a corrosion inhibitor, antioxidant synergist, tetraalkyl lead stabilizer and pre-ignition suppressor. However, when desired, the composition of the present inven-

tion may be used along with additional antioxidant, metal deactivator, additional detergent, dye or other additives incorporated in gasoline for specific purposes. When desired, one or more of these additional additives may be admixed with the composition of the present invention and marketed and used in this manner.

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 phosphate-amine salts of this example comprise 75% by weight of the mixed mono- and diisooctyl acid orthophosphate salts of N-tallow-1,3-diaminopropane and 25% by weight of the mixed mono- and ditiodecyl acid orthophosphate salts of 2-ethylhexyl amine. These salts were each separately prepared by intimately mixing the phosphates and amines in proportions to form the neutral salts and then blending them with xylene solvent to form a final solution containing the active ingredients in a concentration of 50% by weight.

A number of compositions of the salts prepared in the above manner with hexylene glycol were prepared. These various blends were used in evaluations to determine the anti-icing properties of the compositions.

The anti-icing properties were determined in a carburetor icing demonstrating apparatus consisting of a vacuum pump equipped so that cool moisture-saturated air from an ice tower is drawn through a gasoline carburetor. The gasoline sample passes from a fuel reservoir through a flow meter into the carburetor at a rate of 14.4 lb./hr. The air from the ice tower is passed at a flow rate of 14.4 lb./hr. at a temperature of 40° F. The manifold vacuum is 9.5 in. Hg at the start and 12.5 in. Hg at the end of the test. Evaporation of the gasoline in the carburetor further cools the cold moist air, with resulting ice formation on the throttle plate. The time in seconds is measured until a drop of 3 in. Hg vacuum occurs, which indicates stalling conditions.

The results reported below are the average of a number of runs.

The fuel used in this example is a commercial gasoline having an API gravity at 60° F. of 75.2, a Reid Vapor Pressure of 13.3 p.s.i. and a boiling range of 87° to 386° F. A sample of this gasoline without anti-icing additive reached stalling conditions within 5 seconds.

The mixed phosphate-amine salts were prepared in the above manner as a 50% solution in the solvent. However, the data reported in the following examples are on the basis of active ingredients of phosphate-amine salts and polyhydroxy glycol but not including the solvent.

Another sample of gasoline containing 5 parts per million of the mixture of phosphate-amine salts described above (no hexylene glycol) reached stalling conditions in 10.3 seconds.

Another sample of gasoline containing 40 parts per million of hexylene glycol (no phosphate-amine salts) reached icing conditions at 10.6 seconds.

In contrast to the above, another sample of the gasoline containing 5 parts per million of the phosphate-amine salts described above and 40 parts per million of hexylene glycol did not reach icing conditions until 13.8 seconds. This increase in icing time from 10.3 or 10.6 seconds to 13.8 seconds is surprising and due to a synergistic effect. Normally it would be expected that the icing time would be the average of the times obtained when using the individual components separately.

Example II

Another series of evaluations was made in the same manner as described in Example I except that the hexylene glycol was used in a concentration of 60 parts per million. When the hexylene glycol in this concentration without the phosphate-amine salts was used, the icing time was 11.4 seconds. In contrast, the use of the mixture of 5 parts per million of the phosphate-amine salts described

in Example I and 60 parts per million of hexylene glycol, the icing time was increased to 15.9 seconds. Here again the synergistic effect of the mixture is demonstrated.

Example III

In another series of evaluations similar to that described in Example I, the phosphate-amine salts were used in a concentration of 10 parts per million and, without hexylene glycol, increased the icing time from 5 to 11.6 seconds. When hexylene glycol was used in a concentration of 80 parts per million, but without the phosphate-amine salts, the icing time was increased to 12.2 seconds.

In contrast to the above, the use of 10 parts per million of the phosphate-amine salts described in Example I and 80 parts per million of hexylene glycol increased the icing time to 25 seconds. This again demonstrates the synergistic effect obtained by using the anti-icing composition of the present invention.

Example IV

Another series of evaluations was made in the same manner as described in Example I except that a different commercial gasoline was used as the fuel. This gasoline had a boiling range of from 85° to 402° F.

A sample of this gasoline without anti-icing additive had an icing time of 11.6 seconds. The addition of 7.5 parts per million of the phosphate-amine salts described in Example I (no hexylene glycol) increased the icing time to 13.5 seconds. Another sample of the gasoline containing 80 parts per million of the hexylene glycol but no phosphate-amine salts increased the icing time to 12.3 seconds.

In contrast, another sample of the gasoline containing 7.5 parts per million of the phosphate-amine salts described in Example I and 80 parts per million of hexylene glycol increased the icing time to 26 seconds, thus again demonstrating the synergistic effect obtained by using the mixture of the present invention.

Example V

Another series of evaluations was made with the same gasoline described in Example IV. The phosphate-amine salts prepared as described in Example I and hexylene glycol were used in a concentration of 10 parts per million and 40 parts per million respectively. This served to increase the icing time to 17.4 seconds. The phosphate-amine salts when used in a concentration of 10 parts per million, but no hexylene glycol, increased the icing time to 14.1 seconds. When the hexylene glycol was used in a concentration of 40 parts per million, without the phosphate-amine salts, the icing time was increased to only 12.5 seconds.

Example VI

The anti-icing composition of this example consisted of 25% by weight of the phosphate-amine salts prepared as described in Example I, 74.6% by weight of hexylene glycol and 0.4% by weight of glycerol. This composition was incorporated in a concentration of 40.7 parts per million in a commercial gasoline and served to increase the icing time from 9.7 seconds to 13.1 seconds.

When the above composition was used in a concentration of 60 parts per million, the icing time was increased to 14.4 seconds.

Example VII

The anti-icing composition of this example is a blend of 20% by weight of a mixture of 60% by weight of mono- and diisooctyl acid orthophosphate salt of N-tallow-1,3-diaminopropane and 40% by weight of mono- and ditiodecyl acid orthophosphate salt of 2-ethylhexylamine and 80% by weight of 1,6-dihydroxyhexane. The composition is incorporated into a commercial gasoline in a concentration of 60 parts per million and serves to prevent stalling and also serves to maintain cleanliness of the carburetor throttle body.

Example VIII

The anti-icing composition of this example is a mixture of 50% by weight of a mixture of 35% by weight of the diamyl acid orthophosphate salt of N-oleyl-1,3-diaminopropane and 65% by weight of diamyl acid orthophosphate salt of oleylamine and 50% by weight of ethylene glycol. The mixture is prepared as a 50% solution in toluene solvent and is incorporated in a concentration of 50 parts per million into commercial gasoline. The gasoline is improved both in anti-stalling property and in carburetor cleanliness.

We claim as our invention:

1. Synergistic anti-icing composition of from about 5% to about 95% by weight of (1) from about 5% to about 95% by weight of alkyl acid phosphate salt of N-alkyl-diaminoalkane in which the alkyl contains from 3 to about 30 carbon atoms and the alkane contains from 2 to about 12 carbon atoms and from about 5% to about 95% by weight of alkyl acid phosphate salt of alkyl-monoamine in which the alkyl contains from 3 to about 20 carbon atoms and (2) from about 95% to about 5% by weight of polyhydroxy aliphatic alcohol containing from 2 to about 8 carbon atoms and from 2 to 4 hydroxy groups.

2. The composition of claim 1 in which said phosphate salt of N-alkyl-diamino-alkane is a mixture of mono- and ditridecyl acid orthophosphate salts of N-tallow-1,3-diaminopropane.

3. The composition of claim 1 in which said phosphate salt of N-alkyl-diamino-alkane is a mixture of mono-

and dioctyl acid orthophosphate salts of N-tallow-1,3-diaminopropane.

4. The composition of claim 1 in which said phosphate salt of alkyl-monoamine is a mixture of mono- and ditridecyl acid orthophosphate salts of 2-ethylhexyl amine.

5. The composition of claim 1 in which said polyhydroxy alcohol is hexylene glycol.

6. The composition of claim 1 in which said polyhydroxy alcohol is ethylene glycol.

7. The composition of claim 1 incorporated in gasoline to prevent stalling of an internal combustion engine due to icing.

8. The composition of claim 7 incorporated in a concentration of from about 0.0001% to about 0.05% by weight of the gasoline.

References Cited

UNITED STATES PATENTS

2,701,754	2/1955	Hayworth et al.	
2,936,223	5/1960	Lovett et al.	44-77 XR
3,007,782	11/1961	Brown et al.	44-72 XR
3,063,820	11/1962	Chenicek.	
3,228,758	1/1966	Bauer	44-72

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