United States Patent Office 3,573,011
Patented Mar. 30, 1971

3,573,011

GASOLINE COMPOSITION

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No Drawing. Filed Sept. 30, 1968, Ser. No. 763,974
Int. Cl. C10l 1/18
U.S. Cl. 44—66 12 Claims

ABSTRACT OF THE DISCLOSURE

A gasoline motor fuel having improved anti-icing, detergency, anti-rust and ignition control characteristics is obtained by incorporating in the fuel a small amount of a combination of a polyoxypropylene ester and a monocarboxylic acid salt of an N-aliphatic substituted polymethylene diamine. The polyoxypropylene ester has the general formula

$$RCOOC_3H_6O(C_3H_6O)_nC_3H_6OH$$

where R is an aliphatic hydrocarbon radical containing 7 to 29 carbon atoms and n is an integer of 20 to 100. The monocarboxylic acid salt is the salt of a monocarboxylic acid which contains at least 8 carbon atoms in the molecule and an N-aliphatic substituted polymethylene diamine which has the general formula $RNH(CH_2)_xNH_2$ where R is an aliphatic hydrocarbon radical containing 8 to 30 carbon atoms and x is an integer of 2 to 10.

This invention relates to fuels and more particularly to gasoline motor fuels for high compression spark ignition engines.

Spark ignition engines are operated at speeds varying from slow to fast at atmospheric conditions varying from dry to humid at low to high temperatures. When a spark ignition engine is operated at cool, humid atmospheric conditions, using a gasoline fuel having a relatively low 50 percent ASTM distillation point, i.e., below about 235° F., excessive engine stalling is apt to be encountered at idling speeds during the warmup period, especially where engine idling occurs following a period of light load operation. Engine stalling under such conditions has been attributed to the partial or complete blocking of the narrow air passage that exists between the carburetor throat and the carburetor throttle valve during engine idling, by ice particles that deposit upon and adhere to the metal surfaces of the carburetor parts. Such icing of carburetor parts occurs as a result of the condensation of moisture from the air drawn into the carburetor and as a result of the solidification of such condensed moisture. The aforesaid condensation and solidification of moisture are caused by the refrigerating effect of rapidly evaporating gasoline. Accordingly, excessive engine stalling due to carburetor icing occurs as a practical matter only in the instance of gasolines containing a large proportion of relatively high voltatile components. In practice, the problem of engine stalling due to carburetor icing has been found to be serious, under cool, humid atmospheric conditions, in connection with gasolines having a 50 percent ASTM distillation point below about 235° F.

Engine stalling can be caused not only by ice which is deposited in the carburetor but also by other deposits which may form in the carburetor, particularly in the throttle body area of the carburetor. While such other deposits may form under any driving conditions, these deposits are more likely to be formed in the carburetor of an engine operating at idling speeds during a considerable period of the operating time. Such driving conditions are normally encountered in engines of automobiles used to a great extent in driving in heavy traffic of the type encountered in city driving.

As deposits begin to build up in the throttle area of the carburetor, the clearance between the throttle plate and the body wall of the carburetor becomes progressively less. As the clearance is reduced the amount of air passing the throttle plate for a given amount of fuel is also reduced. As a result of a reduced amount of air, the air-fuel mixture introduced into the combustion chamber is richer than it should be for satisfactory engine operation. As deposits continue to build up in the carburetor, rough engine idling is encountered and eventually the engine will stall under idling conditions.

The deposits which are formed in the carburetor may be due in part to the make-up of the motor fuel which is used, but it is believed that the deposits are due, to a greater extent at least, to foreign matter introduced into the carburetor through the air intake system. Air cleaners employed in automotive engines do not appear to effectively remove these contaminants. Major contributors to air contamination are crankcase vapors, exhaust vapors, dust, smoke and the like. The problem with respect to carburetor deposits resulting from air contamination is further aggravated by positive crankcase ventilating systems which are employed in many of the current automotive engines. In engines equipped with a positive crankcase ventilating system, fumes and/or vapors from the crankcase are passed through a metering valve to the air intake system of the engine. While this system helps to cut down on fumes escaping to the atmosphere the system adds to the problem of deposits formed in the carburetor.

Excessive engine stalling whether resulting from the formation of ice or from other deposits in the carburetor is, of course, a source of annoyance due to the resulting increased fuel consumption, battery wear and inconvenience of frequent restarting. It is, therefore, important that any inherent engine stalling characteristics of gasoline fuels due to carburetor icing be reduced substantially and that the formation of other deposits also be reduced. While ice deposits will melt eventually, other deposits which accumulate in the carburetor must either be removed or the carburetor must be replaced before satisfactory engine performance can be obtained.

I have found that engine stalling resulting from carburetor icing and the formation of other deposits in the carburetor of a spark ignition engine can be substantially reduced by incorporating in the gasoline motor fuel a small, deposit inhibiting amount of a combination of (1) an oil-soluble polyoxypropylene ester having the general formula:

$$RCOOC_3H_6O(C_3H_6O)_nC_3H_6OH$$

where R is an aliphatic hydrocarbon radical containing 7 to 29 carbon atoms and n is an integer of 20 to 100 and (2) an oil-soluble monocarboxylic acid salt of an N-aliphatic substituted polymethylene diamine, wherein said monocarboxylic acid contains at least 8 carbon atoms per molecule and wherein said N-aliphatic substituted polymethylene diamine has the general formula:

$$R-\overset{H}{\underset{|}{N}}-(CH_2)_x-NH_2$$

where R is an aliphatic hydrocarbon radical containing about 8 to about 30 carbon atoms and x is an integer from 2 to 10. The present invention, therefore, includes a gasoline motor fuel composition that has been improved in the above-described fashion, and also the method of operating an internal combustion engine under conditions normally tending to form deposits in the carburetor using such improved gasoline composition as the fuel. The monocarboxylic acid salt and the polyoxypropylene ester can be employed in varying proportions with respect to one another. It is generally preferred to add them in weight proportions of about 1:1 to about 2:1, but other propor-

tions can be used, provided that each compound is present in an amount corresponding to at least about 0.001 percent by weight of the gasoline composition. Usually the compounds will be employed in weight ratios varying from about 1:5 to about 5:1.

The exact manner of functioning of the herein disclosed combinations of monocarboxylic acid salt and polyoxypropylene ester has not been definitely determined, and accordingly, the invention is not limited to any particular theory of operation. It may be that such combinations inhibit the formation of deposits in the carburetor by preventing their formation in the first instance. Alternatively, it may be that the herein disclosed combinations of carboxylic acid salt and polyoxypropylene ester function as solubilizing agents for the deposits. However, regardless of the particular mechanism by which the herein disclosed combinations function, it is clear that the components of the combination coact in a unique manner to provide substantially reduced engine stalls resulting from carburetor icing and also reduced deposits in the carburetor of a spark ignition engine.

The polyoxypropylene esters employed in accordance with the present invention are nonvolatile liquids at normal atmospheric conditions and have viscosities of about 200 to about 1000 SUS at 100° F. The molecular weight of the polyoxypropylene ester naturally depends upon the number of propylene oxide molecules in the compound and upon the fatty acid used in forming the ester. In general, the utilizable polyoxypropylene esters will have an average molecular weight of about 1,000 to about 8,000.

Polyoxypropylene esters are available commercially therefore, neither the esters per se nor their method of preparation constitute any portion of the present invention. The polyoxypropylene esters, for example, can be prepared by reacting the desired fatty acid and propylene oxide in mole ratios of about 1:20 to about 1:100 or more, fatty acid to propylene oxide, respectively. The polyoxypropylene ester appears to have a hydroxyl group at one end of the chain and the aliphatic group of the starting acid at the other end. The reaction is believed to be a simple addition wherein the propylene oxide molecules undergo conversion to the corresponding oxypropylene radicals as illustrated by the following equation:

$$RCOOH+n(C_3H_6O)\rightarrow RCOOC_3H_6O(C_3H_6O)_{n-2}C_3H_6OH$$

where R is an aliphatic hydrocarbon radical containing 7 to 29 carbon atoms and n is an integer of 20 to 100. The polyoxypropylene esters suitable for the purposes of this invention include esters of long-chain monobasic fatty acids that contain at least 8, and preferably 12 to 24 carbon atoms per molecule. Esters of fatty acids that contain on the average about 18 carbon atoms per molecule are particularly effective. Either saturated or unsaturated monobasic fatty acid residues containing from 8 to 30 carbon atoms can be employed. Examples of fatty acids that form suitable esters for the purposes of the invention are caprylic acid, pelargonic acid, nonylenic acid, capric acid, decylenic acid, undecylenic acid, lauric acid, myristic acid, palmitic acid, stearic acid, recinoleic acid, oleic acid, linoleic acid, linolenic acid, arachidic acid, behenic acid, erucic acid, brassidic acid, lignoceric acid, carnaubic acid, cerotic acid, octacosoic acid and melissic acid. Specific examples of preferred esters for use in the composition of this invention are polyoxypropylene monolaurate, polyoxypropylene monopalmitate, polyoxypropylene monostearate and polyoxypropylene monooleate. Commercially available mixtures of such esters can also be employed.

The polyoxypropylene ester is employed in the gasoline motor fuel composition of the invention in small amounts ranging from about 0.001 to about 0.01 percent by weight of the gasoline composition but preferably is within the range of about 0.002 to about 0.004 percent by weight. Excellent results have been obtained when the

polyoxypropylene ester was employed in the gasoline in an amount corresponding to about 0.0023 percent by weight based on the weight of the gasoline, i.e., about 6 pounds of polyoxypropylene ester per 1000 barrels of gasoline. While amounts in excess of about 0.01 percent can be employed without deleteriously affecting the other properties of the composition, such larger amounts do not give significantly improved detergency characteristics in combination with the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine. Therefore, for economic reasons, I prefer to use no more of the polyoxypropylene ester than is necessary to give the desired improvement. The polyoxypropylene ester is advantageously used in amounts equal in weight to the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine.

Monocarboxylic acid salts of N-aliphatic substituted polymethylene diamines are available commercially and, therefore, neither such salts nor their method of preparation constitute any portion of the present invention. According to one embodiment, the monocarboxylic acid salt of an N-aliphatic substituted polymethylene diamine is prepared merely by mixing the acid with the diamine in the liquid phase. An exothermic neutralization reaction takes place. The temperature is advantageously maintained between about 100° and 230° F. The reaction product is a salt of the diamine.

In preparinig the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine, it is preferred to admix about 1 to about 2 moles of the acid to each mole of diamine. If more than about 2 moles of the acid is reacted with one mole of the polymethylene diamine, the reaction apparently will not go to completion. In order to avoid an excess of acid, it is usually desirable to employ slightly less than 2 moles of the acid with each mole of the diamine. Inasmuch as a diacid salt is preferred over the mono-acid salt, the amount of acid employed should be substantially more than one mole. The reaction temperature is preferably kept below about 230° F. in order to assure salt formation. If the reaction temperature exceeds about 230° F. for an extended period of time the formation of amides, rather than salts, may become excessive.

The monocarboxylic acid with which the N-aliphatic substituted polymethylene diamine is reacted to form the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine for use in the gasoline motor fuel composition of the invention can be a substantially pure acid such as oleic acid but for reasons of economy is preferably commercially available mixtures. Either saturated or unsaturated aliphatic or alicyclic monocarboxylic acids having at least 8 carbon atoms, preferably about 8 to about 30 carbon atoms per molecule can be employed. By way of example, good results have been obtained with oil-soluble petroleum naphthenic acids. As is known, oil-soluble petroleum naphthenic acids consist principally of mixed alicyclic monocarboxylic acids containing 8 or more carbon atoms per molecule, and are recovered by alkali washing of petroleum distillates such as kerosene, naphtha, gas oil and lubricating distillates and by subsequent acidification of the naphthenic acid salts thus obtained. Analysis of mixed naphthenic acids isolated from lubricating oils indicate that they contain from 14 to 30 carbon atoms and are monobasic acids with an average of 2.6 rings. Such acid mixtures normally possess average molecular weights ranging from about 200 to about 450. Although oil-soluble naphthenic acids derived from petroleum are preferred, the invention also includes the use of oil-soluble synthetic naphthenic acids. Examples of such acids are cyclohexyl acetic, cyclohexyl propionic and cyclohexyl stearic acids.

The invention is not limited to oil-soluble petroleum naphthenic acids, inasmuch as effective results are also obtained with combinations of the polyoxypropylene esters of the class disclosed herein and oil-soluble open-

chain or acyclic, aliphatic monocarboxylic acids containing 8 or more preferably about 8 to about 30 carbon atoms per molecule. Specific examples of preferred acids within this class are 2-ethylhexanoic, oleic acid and stearic acid. Examples of other acids within this class are caprylic, pelargonic, nonylenic, capric, decylenic, undecylenic, lauric, myristic, palmitic, ricinoleic, linoleic, arachidic, behenic, erucic, brassidic, carnaubic, cerotic, octacosoic and melissic acids. Mixtures of long chain fatty acids containing from 8 to 20 carbon atoms per molecule such as can be obtained from the saponification of natural fats and oils are also suitable. Examples of such acids are coconut oil (C_8–C_{18} fatty acids), tallow (C_{14}–C_{18} fatty acids) and soybean oil (C_{16}–C_{20} fatty acids).

Specific examples of the naphthenic acid and C_{18} fatty acid salts of the N-aliphatic substituted polymethylene diamines are: N-hexadecyl-trimethylene diamine naphthenate, N-hexadecyl-trimethylene diamine mono-oleate, N-hexadecyl-trimethylene diamine dioleate, N-octadecyl-trimethylene diamine naphthenate, N-octadecyl-trimethylene diamine mono-oleate, N-octadecyl-trimethylene diamine dioleate, N-tallow-trimethylene diamine naphthenate, N-tallow-trimethylene diamine mono-oleate, N-tallow-trimethylene diamine dioleate, N-soya-trimethylene diamine naphthenate, N-soya-trimethylene diamine mono-oleate, N-soya-trimethylene diamine dioleate, N-coco-trimethylene diamine naphthenate, N-coco-trimethylene diamine mono-oleate and N-coco-trimethylene diamine dioleate. The naphthenic acid salts of the N-aliphatic substituted polymethylene diamines are economically attractive inasmuch as they can be prepared from commercial grades of naphthenic acid The oleic acid salts are also economically attractive inasmuch as they can be prepared from a commercial grade of oleic acid known as Red oil. The oleic acid and naphthenic acid salts are particularly desirable also because of their good organophilic characteristics.

The N-aliphatic substituted polymethylene diamine which is reacted with the monocarboxylic acid has the general formula

$$R-\underset{\displaystyle H}{\overset{|}{N}}-(CH_2)_x-NH_2$$

where R is an aliphatic hydrocarbon radical containing about 8 to about 30 carbon atoms and x is an integer from 2 to 10.

The aliphatic group attached to the nitrogen atom is preferably a higher fatty acid residue, that is, R in the above formula is preferably an alkyl or alkenyl radical obtained from a fatty acid. Either saturated or unsaturated fatty acid residues containing from 8 to 30 carbon atoms are particularly desirable. Fatty acids providing such residues can be obtained from most naturally occurring fats and oils, such as soybean oil, coconut oil, tallow, etc. Good results are obtained when a mixture of compounds in which the aliphatic portion of the molecule varies in chain length corresponding to the various chain lengths of the aliphatic radicals provided by naturally occurring mixtures of the fatty acids.

While the N-aliphatic substituted polymethylene diamines can contain from 2 to 10 methylene groups in the molecule, it is generally preferred to employ those compounds containing 2 to 6 and especially 3 methylene groups. Thus, an especially preferred class of N-aliphatic substituted polymethylene diamines are those having the general formula:

$$R-\underset{\displaystyle H}{\overset{|}{N}}-CH_2-CH_2-CH_2-NH_2$$

where R is an aliphatic hydrocarbon radical containing from 8 to 30 carbon atoms. Specific examples of such N-aliphatic substituted polymethylene diamines are N-octyl trimethylene diamine, N-tetradecyl trimethylene diamine, N-tetradecenyl trimethylene diamine, N-hexadecyl trimethylene diamine, N-eicosyl trimethylene diamine, N-

eicosenyl trimethylene diamine, N-docosyl trimethylene diamine, N-docosenyl trimethylene diamine, N-docosodienyl trimethylene diamine, and N-triacontanyl trimethylene diamine. Within the general class of N-aliphatic substituted trimethylene diamines which I can use, those in which the aliphatic N-substituent is an alkyl or alkenyl group containing at least 12 and preferably from 16 to 20 carbon atoms are considered to form especially effective materials. Examples of the N-aliphatic substituted trimethylene diamines which are considered to form especially effective materials are the N-dodecyl, N-hexadecyl trimethylene diamines, and especially the 18 carbon alkyl-, alkenyl-, and alkadienyl-substituted trimethylene diamines such as the N-octadecyl-, N-octadecenyl-, and N-octadecadienyl trimethylene diamines. Mixtures of N-aliphatic substituted trimethylene diamines such as are formed when the aliphatic N-substituent is derived from mixed fatty acids obtained from naturally occurring fats and oils, form highly effective materials for use in the composition of the invention. In such instances the aliphatic N-substituent is a straight-chain monovalent hydrocarbon radical containing from 8 to 20 carbon atoms. Examples of such mixtures of N-aliphatic substituted trimethylene diamines are N-"tallow" trimethylene diamine, N-"soya" trimethylene diamine and N-"coco" trimethylene diamine, where the respective N-substituents are mixed alkyl and unsaturated alkyl groups derived from animal tallow (C_{14}–C_{18}) fatty acids, soybean (C_{16}–C_{20}) fatty acids, and coconut (C_8–C_{18}) fatty acids.

The N-aliphatic substituted polymethylene diamines can be prepared by various well-known chemical procedures. According to one method, a fatty acid is treated with ammonia in the presence of a suitable solvent and catalyst to obtain the corresponding aliphatic nitrile. The nitrile is then hydrogenated under suitable conditions to obtain the corresponding primary amine. The primary amine is then treated with an aliphatic nitrile such as acrylontirile to obtain the corresponding cyanoalkyl aliphatic amine. The cyanoalkyl aliphatic amine is then hydrogenated to obtain the N-aliphatic substituted polymethylene diamine. According to another method, a polymethylene diamine containing the desired number of methylene groups is reacted with an aliphatic chloride containing the desired number of carbon atoms. Since N-aliphatic substituted polymethylene diamines are well-known in the art and are commercially available, no further discussion of their preparation is considered necessary. Exemplary of commercially available N-aliphatic substituted polymethylene diamines are Duomeen T and Duomeen S (products of Armour and Company) which have the general formula $RNHCH_2CH_2CH_2NH_2$ wherein R is derived from tallow fatty acid ("Duomeen T") and from soya fatty acid ("Duomeen S"), respectively.

The following is a description of a typical preparation of a naphthenic acid salt of N-tallow-trimethylene diamine useful for purposes of the present invention. To 27.6 grams (0.1 mol) of petroleum naphthenic acid having a molecular weight of 276 are added at room temperature, with stirring, 20.0 grams (0.05 mol) of Duomeen T (a product of Armour and Company) which has the general formula $RNHCH_2CH_2CH_2NH_2$ wherein R is derived from tallow fatty acids, said Duomeen T having a molecular weight of about 400. Upon completion of the addition of the diamine to the acid, the temperature of the mixture is about 120° F. The reaction mass is then heated to about 210° to about 230° F. with stirring until a neutral salt is obtained. The salt thus obtained is designated as N-tallow-trimethylene diamine naphthenate.

The monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine is employed in the gasoline motor fuel composition of the invention in small amounts ranging from about 0.001 to about 0.01 percent by weight of the gasoline composition, but preferably is within the range of about 0.002 to about 0.004 percent

by weight. Excellent results have been obtained when the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine was employed in amounts corresponding to about 0.0023 percent by weight based on the weight of the gasoline, i.e., about 6 pounds of the acid salt per 1000 barrels of gasoline. While amounts in excess of about 0.01 percent can be employed without deleteriously affecting the other properties of the composition, such larger amounts do not give significantly improved detergency characteristics in combination with the polyoxypropylene ester. Therefore, for economic reasons, I prefer to use no more of the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine than is necessary to give the desired improvement. The acid salt is advantageously used in amounts equal in weight to the polyoxypropylene ester.

The gasoline fuel composition to which the polyoxypropylene ester and the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine are added and in which these components perform the functions described include substantially all grades of gasoline presently being employed in automotive and internal combustion aircraft engines. Such gasolines comprise a mixture of hydrocarbons which can be obtained by at least one of the petroleum conversion processes including cracking, alkylation, aromatization, cyclization, isomerization, hydrogenation, dehydrogenation, hydroisomerization, polymerization, hydroforming, polyforming, "Platforming" and combinations of two or more such processes, as well as by the Fischer-Tropsch and related processes. Thus, the term "gasoline" is used herein in its conventional sense to include hydrocarbons boiling in the gasoline boiling point range. While current straight-run gasoline has octane numbers too low to qualify as the sole hydrocarbon component of gasoline fuel compositions having desirably high octane numbers, a small amount of straight-run gasoline can be blended with the hydrocarbon mixture obtained by one or more of the designated conversion processes provided the resulting mixture has a motor octane number (leaded) of at least about 85 and a research octane number (leaded) of at least about 95. A preferred gasoline fuel composition comprises a blend of hydrocarbons obtained by catalytic cracking, "Platforming" and alkylation processes.

In addition to the polyoxypropylene ester and the monocarboxylic acid salt of an N-aliphatic substituted polymethylene diamine, the gasoline motor fuel composition of my invention can contain conventional amounts of additives commonly employed in a commercial motor fuel including a tetraalkyl lead, an upper cylinder lubricant, a corrosion and oxidation inhibitor, an alkyl halide lead scavenging agent, an alcoholic anti-stalling agent, a metal deactivator, a dehazing agent, an anti-rust additive, an ignition control agent, a dye and the like. Suitable gasolines may contain up to about 5 cubic centimeters of tetraethyl lead fluid per gallon of gasoline.

When an upper cylinder lubricant is employed it is generally used in an amount of from about 0.25 to about 0.75 percent by volume of the composition, e.g., 0.5 volume percent. This oil should be a light lubricating oil distillate, e.g., one having a viscosity at 100° F. of from about 50 to about 500 Saybolt Universal seconds, e.g., about 100 SUS. Although highly paraffinic lubricating distillates can be used, lubricating distillates obtained from Coastal or naphthenic type crude oils are preferred because of their superior solvent properties. The lubricating oil can be solvent-treated, acid-treated, or otherwise refined.

When an oxidation inhibitor is desired, any of the conventional inhibitors can be utilized. The alkylated phenols, e.g., 2,4,6-tri-tertiary-butylphenol, 2,6-di-tertiary-butyl-4-methylphenol, 2,2-bis(2-hydroxy-3-tertiary-butyl-5-methylphenyl) propane and bis(2-hydroxy-3-tertiary-butyl-5-methylphenyl) methane, because of their hydrocarbon-solubility and water-insolubility characteristics are preferred oxidation inhibitors. Such inhibitors when used are incorporated in the gasoline fuel composition in amounts of from about 0.0007 to about 0.02 percent by weight of the composition, e.g., 0.001 weight percent.

When an ignition control agent is employed such as an organo phosphorus compound, its amount is usually expressed in terms of that which is theoretically required to convert the lead introduced into the fuel in the form of tetraalkyl lead to lead orthophosphate. While improved results can be obtained in some instances with amounts corresponding to less than 0.1 times the amount theoretically required to convert the lead to lead phosphate, it is generally preferred to use an amount equal to about 0.1 to about 0.5 times the amount theoretically required to convert the lead to lead orthophosphate. In view of the fact that the amount of tetraalkyl lead in gasoline varies from one fuel to another, it is difficult to state on a weight basis the amount of a particular compound based upon the weight of the gasoline. However, once knowing the amount of tetraalkyl lead present in the gasoline, it is an easy matter to calculate the amount of the particular compound required on a weight basis. Most gasolines on the market contain between about one and about three cubic centimeters of tetraethyl lead per gallon of gasoline. Based upon such a lead content, the phosphorus compounds may be used in amounts corresponding to about 0.003 to about 0.1 percent by weight based on the weight of the fuel. At any rate, the amount should be sufficient to incorporate between about 0.1 and about 1.0, preferably about 0.1 to about 0.5 times the amount of phosphorus required to convert the lead to lead orthophosphate. Exemplary of the organo phosphorus ignition control agents are: trimethyl phosphate, tributyl phosphate, triphenyl phosphate, tricresyl phosphate, trixylyl phosphate, dimethyl xylyl phosphate, cresyl diphenyl phosphate, dicresyl phenyl phosphate, methyl diphenyl phosphate, methyl dicresyl phosphate, ethyl dicresyl phosphate, diisopropyl phosphate, dibutyl phenyl phosphate, diisoamyl cyclohexyl phosphate, tributoxyethyl phosphate, trimethyl phosphite, triethyl phosphite, tributyl phosphite, triisooctyl phosphite, diethyl amyl phosphite, diisopropyl ethyl phosphite, dimethyl ethyl phosphite, diethyl methyl phosphine, diethyl propyl phosphine, diethyl isoamyl phosphine, tributyl phosphine and the like.

Exemplary of another specific improvement agent which I can use is N,N'-disalicylidene-1:2-diaminopropane as a metal deactivator. The metal deactivator is generally used in small amounts of the order of about 0.0002 to about 0.001 percent by weight based on the fuel composition.

The polyoxypropylene ester and the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine can be incorporated into gasoline compositions in any convenient manner. The respective components can be separately added as such to the gasoline compositions, but it is normally preferred to employ them in the form of a concentrated solution or dispersion in a solvent such as mineral oil, gasoline, naphtha, Stoddard solvent, mineral spirits, benzene, heptane, kerosene or the like. If desired, the respective components of the detergency-anti-icing combination can be incorporated in gasoline fuel compositions in admixture with each other, and/or in admixture with other gasoline improvement agents, such as an antioxidant, an anti-knock agent, an ignition control additive, a dehazing agent, an anti-rust additive, a metal deactivator, an upper cylinder lubricant, a dye and the like. After addition, some circulation of the mixture will normally be desirable to expedite formation of a uniform composition, but this is not absolutely necessary.

Although the combination of the polyoxypropylene ester and the monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine in accordance with the invention is utilized primarily in gasoline for its de-

tergency and anti-icing properties the combination is additionally useful in that it imparts valuable anti-rust characteristics to gasoline when used in anti-icing and detergency-improving amounts. The combination also improves the action of an ignition control agent in reducing engine noise, including preignition, normal knocking and rumble.

As heretofore indicated, the combination of (1) a polyoxypropylene ester and (2) a monocarboxylic acid salt of an N-aliphatic substituted polymethylene diamine included by this invention produces an improvement in the detergent characteristics of gasoline that normally tends to form deposits in the carburetor of an automotive engine. To illustrate the nature of the improvement obtained, there are presented in Table I the results obtained with engine tests made upon a base gasoline motor fuel and the same base gasoline motor fuel containing a combination of polyoxypropylene monooleate and N-tallow trimethylene diamine naphthenate.

Typical properties of the polyoxypropylene monooleate and N-tallow trimethylene diamine naphthenate used in the illustrative examples are as follows:

Properties	Polyoxypropylene monooleate	N-tallow trimethylene diamine naphthenate [1]
Gravity: ° API	9.7	21.6
Specific gravity, 60°/60° F	1.002	0.924
Viscosity, SUV: Sec.:		
At 100° F	996	1,692
At 130° F	515	600
At 150° F	157.8	355
Flash point, P-M: ° F	220	142
Pour point, ° F	−40	−10
Color, ASTM D1500	<1.0	8.0
Neutralization No. ASTM D664, Total Acid No.	1.0	64
Sulfated Ash, ASTM D874, percent	0.002	0.02
Insolubles, ASTM D893, percent:		
n-Pentane	0.01	0.01
Benzene	0.01	0.01
Molecular weight	1,850	

[1] Determinations made in kerosene: (75% by weight N-tallow trimethylene diamine naphthenate; 25% by weight kerosene).

Typical properties of the base gasoline [1] used in the carburetor throat rating test described hereinafter are as follows:

Gravity, ° API	55.6
Specific gravity, 60°/60° F.	0.7563
Doctor, Fed. 520.3.2	Negative (sweet)
Sulfur, ASTM D1266, wt. percent	0.034
Copper strip test, 122° F., 3 hrs.	1.0
Copper dish gum, mg./100 ml.	11.0
Oxidation stability, min.	1440
Bromine No.	28
Knock rating:	
Motor method	90.6
Research method	99.6
TEL, ml./gal.	3.00
Vapor pressure, Reid, lb.	6
Distillation, gasoline:	
Over point, ° F.	105
End point, ° F.	387
10% evap. at ° F.	150
50% evap. at ° F.	234
90% evap. at ° F.	324
Recovery	98.0
Residue	1.1

According to the test procedure followed, the fuel compositions to be tested are burned in a 371 cubic inch, eight cylinder, Oldsmobile engine equipped with an AC Positive Crankcase Ventilating system and a 2-barrel carburetor. In this test, the engine is operated for 100 cycles, each cycle consisting of 36 minutes operation at idle (650±50 r.p.m.) with no load and 12 minutes operation at 1800±50 r.p.m. with a load of 15 brake horsepower. Prior to each test, the crankcase of the engine is flushed with new lubricating oil for a period of ten minutes, a new oil filter is installed and a clean carburetor and a clean positive crankcase ventilation metering valve are installed. The test starts under the idling portion of the cycle. The jacket temperature is maintained at 175°±5° F. during the test period. The air to fuel ratio is set, during idle condition, at 10.5 (±0.3) to 1 at the beginning of each test. The duration of the test is 80 hours. The crankcase of the engine contains a 20/20W, non-detergent oil. At the conclusion of each 80-hour test, the carburetor is removed and examined. The carburetor throat is visually rated, using 0 to denote a clean rating and 22 to signify a maximum deposit rating. The make-up of the fuels tested and the results of the engine tests are shown in Table I.

TABLE I

	Fuel composition	
	A	B
Base gasoline, volume percent	100	100
Added:		
Polyoxypropylene monooleate: lb./1,000 bbl		6.0
N-tallow-trimethylene diamine naphthenate: lb./1,000 bbl		6.0
Engine Tests, Carburetor Throat Rating [1]	21	6

[1] Rating of 0 denotes a clean condition; rating of 22 denotes very heavy deposits.

The data in Table I clearly demonstrate the marked superiority of a gasoline motor fuel composition of the invention (Composition B) over the base gasoline (Composition A).

In order to further illustrate the detergency characteristics of a composition of the invention, a base gasoline was compared with the same gasoline containing 0.0023 percent by weight (6 pounds per 1000 barrels) of each of (1) polyoxypropylene monooleate and (2) N-tallow-trimethylene diamine naphthenate. The comparison was made in accordance with the Laboratory Induction System Deposit (ISD) Detergency Test. This test comprises forming a gum deposit in the test apparatus by evaporating the test fuel in the apparatus by flowing a stream of heated air counter-current to the flow of the fuel. At the completion of the test, the weight of the adhering gum is determined and compared to the reference gasoline (without additives) for an appraisal of the additives' detergency action. The apparatus which is employed is described by C. C. Moore, J. L. Keller, W. L. Kent and F. S. Liggett, "Evaluating Gasoline for Engine Induction System Gums," The Petroleum Engineer, vol. 27, No. 12, pages C19–30 (1955). In conducting the test, a gum deposit is formed on the walls of a steam-jacketed glass U-tube by evaporating two liters of fuel admitted to the system counter-current to a stream of preheated air. The U-tube is then washed with a number of portions of naphtha until a final wash shows no discoloration. The amount of gum adhering to the apparatus is then determined by extracting it with chemically pure acetone, evaporating the acetone extract with filtered, heated air to obtain a gum residue which is then heated in an oven (½ hour at 100–105° C.), cooled and weighed as noted in the published procedure. Results of the determinations using the same gasoline with and without additives are compared in order to evaluate detergency action. Table II summarizes the results obtained in the Laboratory Induction System Deposit (ISD) Detergency Test.

[1] The base gasoline contained 25 pounds (0.01 percent by weight) of 2,6-di-tertiary-butyl-4-methylphenol as an antioxidant and 1.0 pound (0.0004 percent by weight) of N,N'-disalicylidene-1 : 2-diaminopropane as a metal deactivator per 1000 barrels of gasoline.

TABLE II

Composition	Laboratory ISD test, weight of adhering deposits, mg.	Deposit reduction, percent
Base gasoline	149	0
Base gasoline plus (1) and (2)	74	50

NOTE:
(1) 6 pounds of polyoxypropylene monooleate per 1,000 barrels of gasoline.
(2) 6 pounds of N-tallow trimethylene diamine naphthenate per 1,000 barrels of gasoline.

It is evident from the data in Table II that a composition of the invention has improved detergency characteristics over the base gasoline in that the amount of deposits with the gasoline composition of the invention was reduced by 75 milligrams. This corresponds to a 50 percent reduction in deposits.

Gasoline compositions of the invention can be used as fuels for internal combusion engines over an extended period of time without accumulating undesirable deposits in either the carburetor or crankcase ventilating system. Even deposits which have formed from using unimproved gasoline may be effectively removed by using a gasoline motor fuel composition of the invention.

As indicated hereinabove, a composition of the invention also has valuable carburetor anti-icing properties, In order to illustrate the anti-icing properties of a composition obtained in accordance with the invention, a base gasoline having a 50 percent ASTM D86 distillation point of 200° F. was compared with the same gasoline containing 0.0023 percent by weight (6 pounds per 1000 barrels) of each of (1) polyoxypropylene monooleate and (2) N-tallow-trimethylene diamine naphthenate. In order to illustrate the superior anti-icing properties obtained with the combination of the polyoxypropylene monooleate and the N-tallow trimethylene diamine naphthenate, according to the invention, there are also presented in Table III the results obtained with engine tests carried out on the base gasoline containing each of the components alone. The comparison was made in a 225 cubic inch six-cylinder, Plymouth engine operated under a dynamometer load amounting to 15 horsepower on a test stand under cycling conditions in a cold room maintained at 40° F. for a warm-up period of 20 cycles. Each cycle comprises 20 seconds at 1500 r.p.m., followed by an idle for 20 seconds at 525 r.p.m. Air is supplied to the carburetor at ambient temperature (40° F.) and pressure conditions and at 96 percent relative humidity. The engine oil and coolant are circulated through a single heat exchanger to maintain the oil temperature at 48° F. and the coolant at 44° F. The number of engine stalls encountered is observed and reported as stalls per 20 cycles. In the present instance, results were also reported in terms of equivalent gasoline volatility performance. Table III summarizes the results obtained in the above-described carburetor icing engine test.

TABLE III

	Composition			
	C	D	E	F
Base gasoline, volume percent	100	100	100	100
Added:				
Polyoxypropylene monooleate, lb./1,000 bbl		12		6
N-tallow-trimethylene diamine naphthenate lb./1,000 bbl			12	6
Inspection, Road-Load, Cold-Room Engine Test:				
No. of stalls/20 cycles*	11.5	7.8	9.3	6.3
Equivalent gasoline 50% distillation point, ° F	200	216	207	226

*Average of 3 or more runs.

From the results presented in Table III above, it will be seen that the addition to gasoline of 6 pounds of each of polyoxypropylene monooleate and N-tallow trimethylene diamine naphthenate per 1,000 barrels of gasoline reduced the number of stalls during a 20 cycle warm-up period from an average of 11.5 to an average of 6.3 a reduction of more than 45%. The improvement thus obtained is more than would be expected from an observation of the results obtained with the gasoline containing each of the components alone. Stated another way, the stalling characteristics of the base gasoline that as such had a 50 percent ASTM distillation point of 200° F. were reduced by the combination of polyoxypropylene monooleate and N-tallow trimethylene diamine naphthenate so that the stalling characteristics of the resulting gasoline composition were equivalent to those of an uninhibited base gasoline having a 50 percent ASTM distillation point of 226° F. When each of the components were employed alone in the base gasoline, the 50% distillation point obtained with the polyoxypropylene monooleate was 216° F. The 50% distillation point obtained with the N-tallow trimethylene diamine was 207° F.

The anti-icing characteristics of a gasoline composition of the invention are further illustrated according to a fuel line freeze-up static test. In this test, fuel is circulated for 30 minutes through a metal U-tube (0.25-inch O.D., 0.18-inch I.D., 2.25-inch bend diam.). Circulation is then stopped and a 0.20 ml. slug of water (approx. 0.5 inch long in tubing) is injected and allowed to settle to the bottom of the U-tube. A cold bath maintained at a preselected temperature is then placed around the tubing and after temperatures reach equilibrium (5 minutes) circulation is again commenced unless freezing has occurred. The test is repeated at 1° F. decrements of temperature until clogging occurs. Table IV summarizes the results obtained in the above-described fuel line freeze-up static test.

TABLE IV

Composition:	Plugging temperature, ° F.
Base gasoline	7
Base gasoline plus (1) and (2)	2

NOTE.—(1) 6 pounds of polyoxypropylene monooleate per 1,000 barrels of gasoline.
(2) 6 pounds of N-tallow trimethyl diamine naphthenate per 1,000 barrels of gasoline.

It is evident from the data in Table IV that a composition of the invention has improved anti-icing characteristics over the base gasoline in that plugging of the fuel line with the base gasoline occurred at 7° F.; with a composition of the invention, plugging did not occur until a temperature of 2° F. was reached. This corresponds to a 71.4 percent reduction in fuel line freeze-up temperature.

As indicated hereinabove, a composition of the invention also has valuable anti-rust characteristics. In order to illustrate the improved anti-rust characteristics obtained in accordance with the invention, a gasoline containing a combination of (1) polyoxypropylene monooleate and (2) N-tallow-trimethylene diamine naphthenate has been compared with a gasoline containing neither of the components (1) and (2). The comparison was made in accordance with the test described in "ASTM Standards on Petroleum Products and Lubricants," ASTM Designation D665–60 except that the test was conducted for a period of 4 hours at room temperature instead of 24 hours at 140° F. as suggested in the test procedure. Table V summarizes the results obtained in the modified ASTM D665–60 test when comparing a premium grade leaded gasoline with the same gasoline containing 0.0023 percent by weight of polyoxypropylene monooleate (6 lb./1000 bbl.) and 0.0023 percent by weight of N-tallow-trimethylene diamine naphthenate (6 lb./1000 bbl).

TABLE V

Composition	Anti-rust rating after 4 hours at room temperature with the composition in—	
	Distilled water	Sea water
Base gasoline	Heavy Rust	Heavy Rust.
Base gasoline plus (1) and (2)	No Rust	Light Rust.

NOTE:
(1) 6 pounds of polyoxypropylene monooleate per 1,000 barrels of gasoline.
(2) 6 pounds of N-tallow-trimethylene diamine naphthenate per 1,000 barrels of gasoline.

The improved anti-rust characteristics obtained in accordance with my invention are apparent from the summary of data in Table V.

In addition to the detergency, anti-icing and anti-rust characteristics imparted to gasoline by the combination of the polyoxypropylene ester and the monocarboxylic acid salt of the N-aliphatic substituted diamine in accordance with the invention, this combination of additives also improves the action of an ignition control agent in reducing engine noise, including, preignition, normal knocking and rumble.

In order to illustrate the improved ignition control characteristics obtained with fuels of the invention, an octane number requirement test was employed in which the fuel was burned in a commercially available multi-cylinder spark-ignition engine having a compression ratio of 10.25 to 1. The test is intended to assimilate the conditions encountered in city-suburban driving. In this test, the engine is operated on a cycling schedule consisting of one-minute idle at 475 r.p.m., two minutes at 1500 r.p.m. (37 m.p.h.) road load, one-half minute at 2200 r.p.m. (60 m.p.h.) hill climb load and one-half minute at 2200 r.p.m. (60 m.p.h.) road load. Other test conditions are:

Air/fuel ratio at idle—11.5:1
Air/fuel ratio at cruise—14.5:1
Cycle duration—4 min.
Test duration—216 hr.
Total elapsed time—222 hr.
Gasoline required—400 gal.
Water temperature—160° F.
Oil temperature—200° F. (max.)
Carburetor air temperature—85° F.
Induction air humidity—44 grains/lb.
Lube oil required—5 gal.

At the end of each twenty-four hour period during the test, octane requirement and noise determinations are made. After the octane requirement and noise determinations are made, the engine is again placed on the cycling schedule for another twenty-four hours. The octane requirement and noise determinations are continued for nine 24-hour periods or less if an equilibrium octane number requirement appears to have been reached.

The noise determinations are made according to three succesive steps. If noise is encountered in step one, then steps two and three are omitted. If noise is encountered in step two, then only step three is omitted. Noise in this test is intended to include preignition, normal knocking or rumble. The three successive steps of the test are as follows:

(1) At a speed of 1100 r.p.m. the throttle is opened to detent (that is, the rear barrels of the carburetor are just open) at 1-inch Hg intake manifold vacuum.

(2) The engine speed is increased to 1300 r.p.m. at 3-inch vacuum.

(3) The engine is accelerated at 10-inch vacuum from 1300 to 2000 r.p.m. standard spark, and held at this setting for 3 seconds (throttle wide-open at end of 3-second period).

Aural observations are made at steps (1), (2) and (3) and preignition, rumble and knock are recorded.

Ratings are made on the tank fuel (99 research octane number) and the actual noise determined by the use of a set of commercial reference fuels up to an octane number of 113.5. For noise determinations in the range 113.5 to 120, leaded isooctane is used. Octane numbers above 100 are expressed in the approved extension scale, Wiese octane numbers, which are:

$$\frac{\text{Performance No. } 100}{3}+100$$

In addition to the data obtained in the octane number requirement test described above, other octane number requirement data were obtained in accordance with a procedure for determining leaded isooctane-benzene (LIB) ratings. The leaded isooctane-benzene rating, like the average octane number requirement to prevent engine noise, is also a rating of the fuel required to prevent engine noise, including preignition, normal knocking and rumble. The number signifying the LIB rating indicates the lowest percentage of isooctane in a leaded isooctane-benzene mixture that can be burned in the engine without inducing noise. Hence, a low LIB number indicates effective preignition control. In this test the isooctane and benzene each contain 3 cc. of tetraethyl lead per gallon. Inasmuch as there is little if any volume change on mixing benzene with isooctane, the mixture is also assumed to contain 3 cc. of tetraethyl lead per gallon.

Table VI summarizes the average octane number requirement test data and the LIB ratings of the fuel to prevent engine noise including preignition, normal knocking and rumble, when the test engines were operated under the above test procedures with the base gasoline, the base gasoline containing methyl diphenyl phosphate as an ignition control agent and the base gasoline containing methyl diphenyl phosphate in combination with a mixture of polyoxypropylene monooleate and N-tallow-trimethylene diamine naphthenate.

TABLE VI

	Composition						
	G	H	I	J	K	L	M
Base gasoline, vol. percent	100	100	100	100	100	100	100
Added:							
Tetraethyl lead: ml./gal.	3	3	3	3	3	3	3
Methyl diphenyl phosphate, theory		0.1	0.15	0.20	0.1	0.12	0.12
Polyoxypropylene monooleate, lb./1,000 bbl					10	6	10
N-tallow-trimethylene diamine naphthenate, lb./1,000 bbl					10	6	10
Engine tests—Research Octane Number:							
Start	97	97		97	97	97	97
After 216 hours	120+	115	107	105	109	112	102
Avg. last 5 periods	120+	112	107	104	107	110	103
LIB Number:							
Maximum OR	>24	18	11	9	13	15	7
Start	0	0	0	0	0	0	0
After 216 hours	100+	100+	80	80	95	95	70
Avg. last 5 periods	100+	93+	82	77	88	92	73

The data in the foregoing Table VI clearly indicate the improvement in octane requirement increase (ORI) and leaded isooctane-benzene (LIB) ratings obtained with compositions of the invention (Compositions K, L and M). It will be noted that when a small amount of a mixture of polyoxypropylene monooleate and N-tallow-trimethylene diamine naphthenate was added to the base gasoline containing methyl diphenyl phosphate as an ignition control agent, the octane number requirement of the fuel to prevent engine noise was substantially reduced.

The above specific examples have been described in connection with the use of a base gasoline containing a mixture of polyoxypropylene monooleate and N-tallow trimethylene diamine naphthenate. Other illustrative compositions within the scope of the invention are shown in Table VII.

TABLE VII

	Composition				
	N	O	P	Q	R
Base gasoline, vol. percent	100	100	100	100	100
Added, lb./1,000 bbl.:					
Polyoxypropylene monolaurate	5				15
Polyxypropylene monopalmitate		6			
Polyoxypropylene monostearate			8		
Polyoxypropylene monooleate				10	
N-hexadecyl-trimethylene diamine naphthenate	10				
N-octadecyl-trimethylene diamine monooleate				10	
N-tallow-trimethylene diamine naphthenate			8		
N-soya-trimethylene diamine dioleate		6			
N-coco-trimethylene diamine naphthenate					5

The physical characteristics of gasoline are not substantially changed by the addition of a polyoxypropylene ester and a monocarboxylic acid salt of the N-aliphatic substituted polymethylene diamine in accordance with the invention as evidenced by the data in Table VIII. The base gasoline also contained 10 pounds (0.004 percent by weight) of 2,6-di-tertiary butyl-4-methyl phenol as an antioxidant and 1.0 pound (0.0004 percent by weight) of N,N'-disalicylidene-1:2-diaminopropane as a metal deactivator per 1000 barrels of gasoline.

TABLE VIII

Inspections	Base gasoline	Base gasoline plus 6 lb./1,000 bbl. of (1) and 6 lb./1,000 bbl. of (2)
Gravity, ° API	61.5	61.4
Sp., 60°/60° F	0.7332	0.7335
Doctor, Fed. 520.3.2	(3)	(3)
Sulfur, ASTM D1266, Wt. percent	0.009	0.009
Copper strip test, 122° F., 3 hrs	1.0	1.0
Existent gum; mg./100 ml	1	1
Oxidation stability, min	1,440	1,440
Bromine No	19.1	18.7
Knock Rating:		
Motor method	93.8	94.1
Research method	100.8	100.7
TEL. Ml./gal	3.0	3.0
Vapor pressure, Reid, lb	6.30	6.30
Distillation, ASTM:		
Over point, ° F	101	98
End point, ° F	370	369
10% evaporation at ° F	146	151
50% evaporation at ° F	227	226
90% evaporation at ° F	300	303

[1] Polyoxypropylene monooleate.
[2] N-tallow-trimethylene diamine naphthenate.
[3] Negative.

It will be noted from the data in Table VIII that the combination of the polyoxypropylene monooleate and the N-tallow-trimethylene diamine naphthenate had no deleterious effect on the physical characteristics of the gasoline. There is no significant change in the octane number of the gasoline. Likewise, there is no change in the existent gum, the copper corrosivity or the oxidation stability of the fuel. Moreover, it has been found that there is no increase in the emulsion-forming or filter-clogging tendencies of the gasoline.

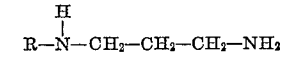
While my invention has been described with reference to various specific examples and embodiments, it will be understood that the invention is not limited to such examples and embodments and may be variously practiced within the scope of the claims hereinafter made.

I claim:

1. A gasoline motor fuel composition comprising a major amount of gasoline normally tending to form deposits in the carburetor of a spark ignition engine and a small amount, sufficient to inhibit the formation of said deposits, of a combination of (1) an oil-soluble polyoxypropylene ester having the general formula:

$$RCOOC_3H_6O(C_3H_6O)_nC_3H_6OH$$

where R is an aliphatic hydrocarbon radical containing 11 to 17 carbon atoms and n is an integer of 20 to 100 and (2) an oil-soluble monocarboxylic acid salt of an N-aliphatic trimethylene diamine, wherein said monocarboxylic acid is selected from the group consisting of oleic acid and a mixture of oil-soluble petroleum naphthenic acids and wherein said N-aliphatic substituted trimethylene diamine has the general formula:



where R is an aliphatic hydrocarbon radical derived from mixed fatty acids selected from the group consisting of coco, tallow and soya fatty acids.

2. The gasoline motor fuel composition of claim **1** wherein said monocarboxylic acid is oleic acid.

3. The gasoline motor fuel composition of claim **1** wherein said monocarboxylic acid is a mixture of oil-soluble petroleum naphthenic acids.

4. A gasoline motor fuel composition comprising a major amount of gasoline normally tending to form deposits in the carburetor of a spark ignition engine and a small amount, sufficient to inhibit the formation of said deposits, of a combination of (1) polyoxypropylene monooleate and (2) N-tallow-trimethylene diamine naphthenate.

5. A gasoline motor fuel composition comprising a major amount of gasoline normally tending to form deposits in the carburetor of a spark ignition engine and a small amount, sufficient to inhibit the formation of said deposits, of a combination of (1) polyoxypropylene monolaurate and (2) N-coco-trimethylene diamine naphthenate.

6. A gasoline motor fuel composition comprising a major amount of gasoline normally tending to form deposits in the carburetor of a spark ignition engine and a small amount, sufficient to inhibit the formation of said deposits, of a combination of (1) polyoxypropylene monosterate and (2) N-tallow-trimethylene diamine naphthenate.

7. A gasoline motor fuel composition comprising a major amount of gasoline normally tending to form deposits in the carburetor of a spark ignition engine and a small amount, sufficient to inhibit the formation of said deposits, of a combination of (1) polyoxypropylene monopalmitate and (2) N-soya-trimethylene diamine dioleate.

8. A gasoline motor fuel composition comprising a major amount of gasoline containing up to about 5 cubic centimeters of tetraethyl lead per gallon of gasoline to produce a gasoline fuel composition having a motor oc-

tane number of at least about 85 and a research octane number of at least about 95; about 0.003 to about 0.1 percent by weight of an organo phosphorus compound, the organo phosphorus compound comprising at least about 0.1 times the theoretical amount required to convert the lead in said tetraethyl lead to lead phosphate; about 0.001 to about 0.01 percent by weight of polyoxypropylene monooleate; and about 0.001 to about 0.01 percent by weight of N-tallow-trimethylene diamine naphthenate.

9. The gasoline motor fuel composition of claim 8 wherein the organo phosphorus compound is methyl diphenyl phosphate.

10. A gasoline motor fuel composition comprising a major amount of gasoline containing about 1 to about 3 cubic centimeters of tetraethyl lead per gallon of gasoline to produce a gasoline fuel composition having a motor octane number of at least about 85 and a research octane number of at least about 95; about 0.003 to about 0.1 percent by weight of methyl diphenyl phosphate, the methyl diphenyl phosphate comprising at least about 0.1 times the theoretical amount required to convert the lead in said tetraethyl lead to lead phosphate; about 0.002 to about 0.004 percent by weight of polyoxypropylene monooleate; and about 0.002 to about 0.004 percent by weight of N-tallow-trimethylene diamine naphthenate.

11. A gasoline motor fuel composition comprising a major amount of gasoline containing about 1 to about 3 cubic centimeters of tetraethyl lead per gallon of gasoline to produce a gasoline fuel composition having a motor octane number of at least about 85 and a research octane number of at least about 95; about 0.003 to about 0.1 percent by weight of methyl diphenyl phosphate, the methyl diphenyl phosphate comprising at least about 0.1 times the theoretical amount required to convert the lead in said tetraethyl lead to lead phosphate; about 0.002 to about 0.004 percent by weight of polyoxypropylene monooleate; about 0.002 to about 0.004 percent by weight of N-tallow-trimethylene diamine naphthenate; about 0.0007 to about 0.02 percent by weight of 2,6-di-tertiary-butyl-4-methylphenol; and about 0.0002 to about 0.001 percent by weight of N,N'-disalicylidene-1:2-diaminopropane.

12. A gasoline motor fuel composition comprising a major amount of gasoline containing about 3 cubic centimeters of tetraethyl lead per gallon of gasoline to produce a gasoline fuel composition having a motor octane number of at least about 90 and a research octane number of at least about 99; about 0.12 times the theoretical amount of methyl diphenyl phosphate required to convert the lead in said tetraethyl lead to lead phosphate; about 0.0023 percent by weight of polyoxypropylene monooleate; about 0.0023 percent by weight of N-tallow-trimethylene diamine naphthenate; about 0.001 percent by weight of 2,6-di-tertiary-butyl-4-methylphenol; and about 0.0004 percent by weight of N,N'-disalicylidene-1:2-diaminopropane.

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PO-1050
(5/69)

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,573,011 Dated March 30, 1971

Inventor(s) Gardner E. Gaston

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 14, the last 9 lines of Table VI starting with "Engine tests-Research Octane Number:" the items should read

--Engine Tests

Research Octane Number							
Start	97	97	97	97	97	97	97
After 216 hours	120+	115	107	105	109	112	102
Avg. Last 5 Periods	120+	112	107	104	107	110	103
Maximum ORI	24	18	11	9	13	15	7
LIB Number:							
Start	0	0	0	0	0	0	0
After 216 hours	100+	100+	80	80	95	95	70
Avg. Last 5 Periods	100+	93+	82	77	88	92	73--

Column 16, line 63, "monosterate" should read --monostearate--.

Signed and sealed this 20th day of July 1971.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.
Attesting Officer

WILLIAM E. SCHUYLE
Commissioner of Pa