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MOTOR FUEL CONTAINING SYNERGISTIC OCTANE APPRECIATOR

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This invention relates to a hydrocarbon fuel composition of high octane rating. More specifically, it involves the discovery that the octane rating of leaded gasoline fuels is substantially improved by the addition of synergistic additive mixtures of hydrocarbyl monocarboxylic acids or tertiary alkyl esters thereof with low molecular weight aliphatic alcohols.

The recent increases in compression ratios of automobile engines have placed a severe strain on petroleum refiners to produce fuels having the octane rating demanded by these engines. Premium fuels at the present time have research octane ratings between 97 and 100 and it has been predicted that premium fuels will require octane ratings between 105 and 110 five years from now in order to satisfy the octane requirements of the high compression automotive engines predicted for that date. In order to produce premium fuels of octane ratings of 97 and above, it has been necessary for refiners to rely heavily on catalytic refining operations such as fluid catalytic cracking, catalytic reforming, alkylation and catalytic isomerization.

Catalytic cracking and catalytic reforming, which are the most widely used refining operations in the production of high octane fuels, produce substantial quantities of aromatics; catalytic cracking also produces a substantial amount of olefins. It is well known that olefins and aromatics, although possessing high octane ratings have a poorer response to organo-lead compounds such as tetraethyl lead than saturated aliphatic gasoline components. Accordingly, as the aromatic and olefinic content of the gasolines have increased to meet the octane levels required by modern automotive high compression engines, the lead response of the resulting fuels has diminished. Stated another way, the octane increment obtainable by the addition of an organo-lead compound decreases as the aromatic and olefin contents of the base fuel increase.

In my commonly assigned copending application Serial No. 689,466, filed October 11, 1957, it is disclosed that hydrocarbyl monocarboxylic acids substantially raise the octane rating of a motor fuel containing an organo-lead anti-knock agent and a substantial concentration of high octane components which may be aromatic hydrocarbons, olefinic hydrocarbons or mixtures thereof. In another commonly-assigned copending application Serial No. 699,944, filed December 2, 1957, in the names of George W. Eckert, Howard V. Hess and Edwin C. Knowles, it is disclosed that t-alkyl esters of hydrocarbyl monocarboxylic acids have a similar octane appreciating action on leaded fuels of this composition.

Low molecular weight aliphatic alcohols have long been used as components of gasoline fuels, methyl, ethyl, isopropyl and butyl alcohols are known to possess high octane ratings and their use per se and as components of high octane gasoline has been suggested in many patents and publications. The recommended concentration of aliphatic alcohol for improving the octane rating of gasoline blends ranges from about 5 to about 50 or more volume percent of the total fuel. The use of small concentrations of low molecular weight aliphatic alcohols, that is, of the order of 0.2 to 2.0 volume percent has been widely practiced in recent years to impart anti-icing, anti-stalling properties to gasoline.

The subject invention involves the discovery that the octane rating of leaded motor fuels containing a substantial concentration of high octane aromatic and/or olefinic components is markedly improved by the addition of a small amount of a mixture of a hydrocarbyl monocarboxylic acid or a tertiary alkyl ester thereof and an aliphatic alcohol.

The high octane hydrocarbon motor fuels of this invention comprise high octane components including a substantial concentration of aromatic hydrocarbons, olefinic hydrocarbons or mixtures thereof, an organo-lead anti-knock agent, a hydrocarbyl monocarboxylic acid or a tertiary alkyl ester thereof in a concentration of at least 0.1 volume percent and a low molecular weight aliphatic alcohol in a concentration of at least 0.2 volume percent of the fuel.

This invention also is directed to a synergistic octane appreciating mixture comprising 1–50 parts of hydrocarbyl monocarboxylic acid, a tertiary alkyl ester or mixture thereof and 2–100 parts of aliphatic alcohol for use in leaded fuels containing the prescribed aromatic and/or olefin content. This synergistic octane appreciating additive advantageously comprises 2–15 parts of monocarboxylic acid or t-alkyl ester and 5–20 parts low molecular weight aliphatic alcohol.

The afore-identified copending applications disclose that the action of monocarboxylic acids and t-alkyl esters in raising the octane rating of gasoline is characterized by several unusual features. First, these agents appear to be ineffective in raising the octane rating of gasolines unless an organo-lead anti-knock agent, normally tertaethyl lead, is a component of the gasoline mixture. Second, equal concentrations of these agents cause a greater octane improvement above the 100 octane level than below the 100 octane level. Third, monocarboxylic acids and t-alkyl esters appear to have little effect on the octane rating of a gasoline consisting essentially of saturated aliphatic hydrocarbons even though an organo-lead anti-knock agent is present. The synergistic octane appreciators comprising a monocarboxylic acid or a tertiary alkyl ester and an aliphatic alcohol is characterized by the same properties and accordingly is effective in a base fuel containing an organo-lead anti-knock agent and a substantial concentration of aromatics and/or olefins.

The novel fuel compositions of this invention have a minimum concentration of aromatic and/or olefin components of about 10 volume percent. The aromatic and/or olefin components of the motor fuel of the invention can constitute as high as 100 volume percent thereof but usually fall between 20 and 80 volume percent. A minimum 10 percent concentration appears to be necessary for the synergistic action of a monocarboxylic acid or its t-alkyl ester and an aliphatic alcohol in improving the octane rating.

The aromatic components of the motor fuel of the invention are generally supplied by catalytic reforming or catalytic cracking operations. Catalytic reformate is particularly high in aromatics. The olefin components of the motor fuel of the invention are derived either from thermal cracking, catalytic cracking or polymerization.

The organo-lead reagent necessary for the action of the synergistic octane improver is a tetraalkyl lead compound of the class known to possess anti-knock action. Tetraethyl lead is practically universally used as an anti-knock agent but other tetraalkyl lead compounds such as tetramethyl lead, tetrabutyl lead, tetraamyl lead, tetrapropyl lead, etc. are known to possess anti-knock properties and may be used in the fuel compositions of the invention.

The tetraethyl lead mixtures commercially available for automotive use contain an ethylene chloride-ethylene bromide mixture as a scavenger for removing lead from the combustion chamber in the form of volatile lead halides. Tetraethyl lead fluid denotes the commercial product which comprises tetraethyl lead, ethylene chloride and ethylene bromide, the latter two reagents being present in 1.0 theory and 0.5 theory, respectively, theory denoting the stoichiometric amount required for reaction with the lead content of the tetraethyl lead.

The organo-lead reagent is present in the fuel compositions of the invention in concentrations between 0.5 ml. per gallon up to the statutory limit of organo-lead reagent concentration which, at the present time, is 3 ml. per gallon in the case of automotive fuel and 4.6 ml. per gallon in the case of aviation fuel. The usual concentration of tetraethyl lead is between 1 and 3 ml. per gallon in automotive gasoline and 2–4.6 ml. per gallon in aviation gasoline.

The monocarboxylic acid or ester component of the synergistic additive of this invention has the general formula: RCOOR′ wherein R is hydrogen or a hydrocarbyl radical containing 1–29 carbon atoms and R′ is H or a tertiary alkyl group containing 4–18 carbon atoms. In the above general formula, R is advantageously an aliphatic, cycloaliphatic, or aryl radical containing 1–8 carbon atoms; if R′ is a t-alkyl group it advantageously contains 4–12 carbon atoms.

Hydrocarbyl monocarboxylic acids which form synergistic octane appreciators with aliphatic alcohols are the following: acetic acid, formic acid, propionic acid, 2-ethylhexanoic acid, lauric acid, stearic acid, 2-ethylbutyric acid, cumic acid, benzoic acid, cyclohexane carboxylic acid, oleic acid and mixtures thereof. Particularly preferred acids for use in the synergistic octane appreciator of this invention are acetic acid, 2-ethylhexanoic acid, propionic acid, benzoic acid, cyclohexane carboxylic acid and hexanoic acid.

Tertiary alkyl esters effective as synergistic octane appreciators with aliphatic alcohols in the fuel compositions of the invention are the following: t-butyl acetate, t-butyl formate, t-amyl propionate, t-amyl caproate, t-amyl heptanoate, t-octyl pelargonate, t-octyl caprate, t-butyl laurate, t-buytl myristate, t-amyl palmitate, t-nonyl stearate, t-amyl behenate, t-dodecyl butyrate, t-amyl hexanoate, t-dodecyl o-cumate, t-dodecyl benzoate, t-amyl cyclohexane carboxylate, t-octyl cinnamate, t-amyl phenyl acetate, t-butyl oleate, t-butyl ester of Snodotte acids (hydrogenated fish oil acids comprising mainly C_{10} to C_{22} acids) and t-butyl esters of coconut fatty acids (comprising mainly a mixture of C_{12} to C_{16} fatty acids).

The preferred tertiary alkyl esters used in the fuel compositions of the invention are derived from aliphatic and aromatic monocarboxylic acids containing 1–9 carbon atoms and from a tertiary alkyl radical containing 4–12 carbon atoms. Preferred tertiary alkyl esters are t-butyl 2-ethylhexanoate, t-butyl benzoate, t-amyl acetate, t-amyl propionate, t-octyl benzoate, t-butyl hexanoate, t-dodecyl hexanoate, t-butyl acetate and t-butyl propionate.

The monocarboxylic acid or t-alkyl ester must be present in the leaded aromatic and/or olefin-containing compositions of the invention in a minimum concentration of 0.1 volume percent before a synergistic octane appreciation is realized. With acid or ester concentrations below 0.1 volume percent, neither octane improvement per se nor synergistic action with alcohols is obtained in leaded gasoline of prescribed composition. The preferred concentration of acid or ester in the synergistic additive falls between 0.2 and 1.5 volume percent with maximum results generally being obtained at a concentration level of about 0.75 volume percent. Although acid or ester concentrations as high as 5 volume percent can be incorporated in the fuel compositions, economic considerations preclude the use of such high concentrations. In addition, it appears there is a fall-off in octane improvement after acid or ester concentration exceeds about 1.5 volume percent.

The aliphatic alcohol component of the synergistic anti-knock additive of this invention has the general formula: ROH wherein R is an aliphatic hydrocarbyl radical containing 1–10 carbon atoms. The preferred alcohols for

use in the synergistic anti-knock additive of the invention contain 1–6 carbon atoms since the lower alcohols not only display a maximum synergistic octane appreciating action with the t-alkyl esters but they are effective anti-icing agents. Examples of alcohols which react synergistically with t-alkyl esters are methyl alcohol, ethyl alcohol, isopropyl alcohol, n-propyl alcohol, isobutyl alcohol, secondary butyl alcohol, t-butyl alcohol, isoamyl alcohol, n-hexyl alcohol, 2-ethylhexanol, isooctyl alcohol and t-amyl alcohol.

Although minimum concentration of alcohol for synergistic octane improvement is set at 0.2 volume percent, the preferred alcohol concentration employed in the synergistic anti-knock additive of the invention falls between about 0.5 and 2.0 volume percent. Although it has been found that concentrations as high as 10 volume percent alcohol can be used in synergistic combination with acids or t-alkyl esters, alcohol concentrations above about 3.0 volume percent are uneconomic.

In Table I there is shown a synergistic octane appreciating action of mixtures of tertiary alkyl esters and methyl alcohol in a gasoline having a Research Octane Number (RON) of 105. The base fuel contained 3 cc. of tetraethyl lead (TEL) per gallon and comprised approximately 10 percent n-butane, 40 percent isobutane-isobutylene alkylate, 10 percent pentenes from fluid catalytically cracked naphtha and 40 percent heavy platformate. By Fluorescent Indicator Analysis (FIA) the base fuel had an aromatic content of approximately 35 percent and an olefin content of approximately 6 percent; its IBP was 90° F. and its end point was 367° F.

TABLE I

Units Improvement in RON With Mixtures of Tertiary Alkyl Esters and Methyl Alcohols in 105 Octane Gasoline

Ester concentration vol. percent	Concentration of methyl alcohol, volume percent								
	0.0	0.25	0.5	1.0	2.0	3.0	4.0	5.0	7.5
No Ester	-----	0.0	0.0	0.1	0.2	0.9	2.1	2.1	3.0
t-Butyl Acetate:									
0.5%	1.7	-----	-----	2.6	-----	-----	-----	-----	-----
0.75%	1.8	2.1	2.7	3.2	3.2	3.7	4.0	4.7	5.4
t-Butyl Propionate:									
0.75%	1.7	-----	-----	2.5	-----	-----	-----	-----	-----
T-Amyl Acetate:									
0.75%	1.9	-----	-----	2.4	-----	-----	-----	-----	-----

The data in Table I show clearly the synergistic octane appreciating action of mixtures of methyl alcohol and t-alkyl esters in leaded high octane gasoline having the prescribed aromatic and/or olefin content. It will be noted that methyl alcohol in amounts up to about 2.0 volume percent, has substantially no octane appreciating action. As the concentration of methyl alcohol increases from 3 to about 7.5 volume percent the octane appreciating action of the alcohol alone rises from about 1 RON unit to 3 RON units. It is significant that a synergistic octane appreciating action is obtained with mixtures of t-alkyl esters and methyl alcohol throughout the whole range of alcohol concentration.

The synergistic action of the t-alkyl esters with methyl alcohol is exemplified in Table I by the fact that a mixture of 0.75 percent t-butyl acetate and 1.0 percent methyl alcohol appreciates the octane rating of 105 octane gasoline by 2.5 units whereas only a 1.9 unit improvement would be expected if the additives acted independently. Similarly, a mixture of 0.75 percent t-butyl acetate and 2.0 percent methyl alcohol effected a 3.2 unit improvement whereas only a 2.0 unit octane improvement would have been obtained if the additives acted independently.

In Table II there is shown the octane appreciating action of t-alkyl esters with C_2 to C_6 aliphatic alcohols.

The 105 octane base fuel employed in obtaining the data shown in Table II had the same composition as the base fuel employed in Table I.

TABLE II

Units Improvement in RON With Mixtures of t-Butyl Esters and Aliphatic Alcohols

Additive:	Increase in RON
0.75% t-butyl acetate	1.8
0.75% t-butyl propionate	1.7
1.0% ethyl alcohol	0.0
1.0% isopropyl alcohol	0.0
1.0% t-butyl alcohol	0.0
1.0% sec-butyl alcohol	0.0
1.0% isobutyl alcohol	0.0
1.0% n-butyl alcohol	0.0
2.0% 2-ethylbutanol	0.0
0.75% t-butyl acetate+1.0% ethyl alcohol	2.9
0.75% t-butyl acetate+1.0% isopropyl alcohol	3.0
0.75% t-butyl acetate+1.0% sec-butyl alcohol	2.5
0.75% t-butyl acetate+1.0% isobutyl alcohol	2.8
0.75% t-butyl acetate+1.0% n-butyl alcohol	2.7
0.75% t-butyl acetate+2.0% 2-ethylbutanol	2.6
0.75% t-butyl propionate+1.0% isopropyl alcohol	2.5
0.75% t-butyl acetate+1.0% t-butyl alcohol	2.7

The synergistic action of t-alkyl esters and methyl alcohol in regular and premium gasolines is shown in Table III. Base Fuel A was a premium grade gasoline containing 3 cc. of TEL per gallon and comprising approximately 50% saturates, 41% aromatics and 9% olefins by FIA method; base fuel A which had in IBP of 89° F. and an end point of 372° F. had a leaded RON of 100.7.

Base fuel B was a regular grade gasoline containing 3 cc. of TEL per gallon and comprising approximately 56% saturates, 22% olefins and 22% aromatics by FIA method; base fuel B which had an IBP of 92° F. and an end point of 367° F., had a leaded RON of about 93.0.

TABLE III

Units Improvement in RON With Mixtures of t-Butyl Acetate and Methyl Alcohol

Additive	Increase in RON	
	Base fuel A	Base fuel B
0.75% t-butyl acetate	1.1	0.2
1.0% methyl alcohol	0.0	0.0
0.75% t-butyl acetate+1.0% methyl alcohol	1.5	0.8

The data in Table III show clearly the synergistic octane appreciating action of mixtures of t-butyl acetate and methyl alcohol in regular and premium grade gasoline. It is significant that a greater increase in RON is obtained with the synergistic mixture in the premium fuel than in the regular fuel. Comparison with the results shown in Tables I and II indicate that an even greater increase in RON is obtained with the synergistic mixture when the octane level reaches 105. This property of the synergistic mixture is akin to the action of monocarboxylic acids and its t-alkyl esters as disclosed in the afore-identified copending applications Serial Nos. 689,466 and 699,944.

In Table IV there is shown the synergistic octane appreciating action of mixtures of hydrocarbyl monocarboxylic acids and alcohols in the 105 octane base fuel employed in Table I.

TABLE IV

Units Improvement in RON With Mixtures of Monocarboxylic Acids and Aliphatic Alcohols in 105 Octane Gasoline

Additive:	Increase in RON
1.0% methyl alcohol	0.1
1.0% isopropyl alcohol	0.0
1.0% t-butyl alcohol	0.0
0.5% 2-ethylhexanoic acid	2.7
0.3% propionic acid	2.6
0.5% benzoic acid	3.4
0.5% 2-ethylhexanoic acid+1.0% methyl alcohol	3.5
0.3% propionic acid+1.0% methyl alcohol	3.1
0.3% propionic acid+1.0% isopropyl alcohol	4.2
0.5% benzoic acid+1.0% t-butyl alcohol	3.8

The data in Table IV demonstrate the synergistic octane appreciating action of mixtures of hydrocarbyl monocarboxylic acids and aliphatic alcohols in a leaded gasoline containing the prescribed aromatic and/or olefin content.

This invention is an important advance in the technology of high octane fuels since it provides means whereby the significant octane appreciating action of t-alkyl esters and hydrocarbyl monocarboxylic acids on leaded fuels of prescribed compositions is significantly enhanced using readily available reasonably priced materials which are already widely used as gasoline components for their anti-icing function. It is noteworthy that the fuels of this invention containing a synergistic octane appreciating mixture of a hydrocarbyl monocarboxylic acid or a t-alkyl ester thereof with a low molecular weight aliphatic alcohol are characterized by the excellent anti-stalling and anti-icing properties as a result of the presence therein of low molecular weight alcohols.

The selectivity of the synergistic octane appreciating action of mixtures of t-alkyl esters with alcohols was demonstrated by the fact that mixtures comprising 0.75 percent t-butyl acetate and various other oxygenated hydrocarbon derivatives gave the same or poorer octane appreciation than that obtained with 0.75 percent t-butyl acetate alone. Table III lists the oxygenated hydrocarbons which display no synergistic octane appreciating action with t-butyl acetate.

TABLE V

	Percent
Acetone	0.1 to 2.0
Methylethyl ketone	1.0
Mesityl oxide	1.0
Acetophenone	1.0
Diethyl ether	1.0
Morpholine	1.0
Propylene oxide	0.5
Dioxane	0.5

Obviously, many modifications and variations of the invention as hereinbefore set forth may be made without departing from the spirit and scope thereof and, therefore, only such limitations should be imposed as are indicated in the appended claims.

I claim:

1. A hydrocarbon fuel in the gasoline boiling range containing an alkyl lead anti-knock agent, high octane components selected from the group consisting of olefinic hydrocarbons, aromatic hydrocarbons and mixtures thereof in a concentration of at least 10 volume percent of said fuel and a synergistic additive combination of (1) an acidic compound selected from the group consisting of hydrocarbyl monocarboxylic acids containing 1 to 30 carbon atoms, their tertiary alkyl esters and mixtures thereof and (2) a low molecular weight aliphatic alcohol of the general formula: ROH wherein R is an aliphatic hydrocarbyl radical containing 1–10 carbon atoms, said acidic compound being present in a con-

centration between 0.1 and 5.0 volume percent of said fuel and said alcohol being present in a concentration between 0.2 and 5.0 volume percent of said fuel said combination effecting a substantial improvement of the octane rating of said organo-lead-containing hydrocarbon fuel.

2. A hydrocarbon fuel according to claim 1 in which said acidic compound has the general formula: RCOOR' wherein R is selected from the group consisting of hydrogen and hydrocarbyl radicals containing 1–29 carbon atoms, and R' is selected from the group consisting of hydrogen and a t-alkyl group containing 4–18 carbon atoms.

3. A hydrocarbon fuel according to claim 1 in which said alkyl lead anti-knock agent is present in a concentration between 0.5 and 4.6 cc. per gallon.

4. A hydrocarbon fuel in the gasoline boiling range containing an alkyl lead anti-knock agent in a concentration of at least 0.5 cc. per gallon, high octane components selected from the group consisting of olefinic hydrocarbons, aromatic hydrocarbons, and mixtures thereof, in a concentration of at least 10 volume percent of said fuel, a synergistic additive combination of (1) an acidic compound having the general formula: RCOOR' wherein R is a hydrocarbyl radical containing 1–8 carbon atoms and R' is selected from the group consisting of hydrogen and a t-alkyl group containing 4–12 carbon atoms, and (2) a low molecular weight aliphatic alcohol having the general formula: ROH wherein R is an aliphatic hydrocarbyl radical containing 1–6 carbon atoms, said acidic compound being present in a concentration between 0.1 and 5.0 volume percent and said alcohol being present in a concentration between 0.2 and 5.0 volume percent.

5. A hydrocarbon fuel according to claim 4 in which said acidic compound is present in a concentration of 0.2 to 1.5 volume percent and said alcohol is present in a concentration of 0.5 to 2.0 volume percent.

6. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-butyl acetate and methyl alcohol.

7. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-butyl propionate and methyl alcohol.

8. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-butyl acetate and isopropyl alcohol.

9. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-butyl acetate and ethyl alcohol.

10. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-amyl acetate and methyl alcohol.

11. A hydrocarbon fuel according to claim 4 in which said additive combination comprises t-butyl acetate and t-butyl alcohol.

12. A hydrocarbon fuel according to claim 4 in which said additive combination comprises propionic acid and methyl alcohol.

13. A hydrocarbon fuel according to claim 4 in which said additive combination comprises propionic acid and isopropyl alcohol.

14. A hydrocarbon fuel according to claim 4 in which said additive combination comprises 2-ethylhexanoic acid and methyl alcohol.

15. A hydrocarbon fuel according to claim 4 in which said additive combination comprises benzoic acid and t-butyl alcohol.

16. A synergistic octane appreciating additive for leaded fuels containing a substantial concentration of high octane components selected from the group consisting of aromatic and olefinic hydrocarbons and mixtures thereof consisting of 1–50 parts by volume of an acidic compound having the general formula: RCOOR' wherein R is selected from the group consisting of hydrogen and hydrocarbyl radicals containing 1–29 carbon atoms and R' is selected from the group consisting of hydrogen and a t-alkyl group containing 4–18 carbon atoms and 2–100 parts by volume of a low molecular weight aliphatic alcohol of the general formula: ROH wherein R is an aliphatic hydrocarbyl radical containing 1–10 carbon atoms.

17. A synergistic additive combination according to claim 16 comprising 2 to 15 parts by volume of said acidic compound and 5 to 20 parts by volume of said alcohol.

18. A synergistic octane appreciating additive according to claim 16 in which said acidic compound has the general formula: RCOOR' wherein R is a hydrocarbyl radical containing 1–8 carbon atoms and R' is selected from the group consisting of hydrogen and a t-alkyl group containing 4–12 carbon atoms and said alcohol has the general formula: ROH wherein R is an aliphatic hydrocarbyl radical containing 1–6 carbon atoms.

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