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(54) **Compositions for control of deposits, exhaust emissions and/or fuel consumption in internal combustion engines.**

(57) This invention relates to fuel additives and fuels containing such additives for minimizing induction system deposits and for controlling exhaust emissions from an internal combustion engine wherein the fuel additives contain at least the following components :

(a) at least one fuel-soluble succinimide detergent/dispersant ;

(b) a fuel-soluble mineral oil having a viscosity in the range of 10 to 13 cSt at 100°C ;

(c) unhydrogenated poly- α -olefin oligomer having a viscosity in the range of 5 to 10 cSt at 100°C ; and

(d) aromatic hydrocarbon boiling in the range of 160 to 300°C and having a viscosity in the range of 1.4 to 2.0 cSt at 25°C ;

wherein the weight ratio of (d) : (a) is at least 0.5 : 1, the weight ratio of (c) : (a) is in the range of 0.1 : 1 to 3 : 1, the weight ratio of (b) : (c) is in the range of 1 : 1 to 8 : 1, and wherein the weight ratio of (b) + (c) : (a) is in the range of 1.5 : 1 to 2.5 : 1.

This invention relates to fuel additive concentrates, and liquid hydrocarbon fuels of enhanced performance characteristics.

An objective of this invention is to provide fuel additive concentrates and fuel compositions that can minimize induction system deposits in internal combustion engines. Another objective is to provide additive combinations and fuels containing them that are capable of controlling exhaust emissions from internal combustion engines, especially those that have accumulated deposits from use of other fuel compositions. A further objective is to provide compositions that can be used to decrease total exhaust emissions and/or fuel consumption in internal combustion engines, especially those in which deposits have accumulated by virtue of use of other fuel compositions.

In accordance with this invention, it has been found possible to achieve all of the foregoing objectives by use of particular additive components in particular relative proportions to each other in a hydrocarbonaceous fuel.

More particularly, an embodiment of this invention is a fuel additive concentrate which comprises at least the following components:

- (a) at least one fuel-soluble succinimide detergent/dispersant;
- (b) a fuel-soluble mineral oil having a viscosity in the range of 10 to 13 cSt at 100°C;
- (c) unhydrogenated poly- α -olefin oligomer having a viscosity in the range of 5 to 10 cSt at 100°C; and
- (d) aromatic hydrocarbon boiling in the range of 160 to 300°C and having a viscosity in the range of 1.4 to 3.0 cSt at 25°C;

wherein the weight ratio of (d):(a) is at least 0.5:1, the weight ratio of (c):(a) is in the range of 0.1:1 to 3:1, the weight ratio of (b):(c) is in the range of 1:1 to 8:1, and wherein the weight ratio of (b)+(c):(a) is in the range of 1.5:1 to 2.5:1. Preferred are compositions wherein the weight ratio of (b):(c) is in the range of 3:1 to 7:1; the weight ratio of (d):(a) is in the range of 0.7:1 to 20:1; and the weight ratio of (b)+(c):(a) is in the range of 1.7:1 to 2.2:1. Still more preferred are compositions wherein the weight ratio of (b):(c) is in the range of 5:1 to 7:1; the weight ratio of (d):(a) is in the range of 0.7:1 to 4:1; and the weight ratio of (b)+(c):(a) is in the range of 1.8:1 to 2.1:1. Especially preferred are concentrates wherein the weight ratio of (b):(c) is in the range of 5.5:1 to 6.5:1; the weight ratio of (d):(a) is in the range of 0.9:1 to 1.5:1; and the weight ratio of (b)+(c):(a) is in the range of 1.8:1 to 2.1:1. Any of the foregoing concentrates desirably contain at least one fuel-soluble antioxidant, or at least one fuel-soluble demulsifying agent, or at least one fuel-soluble corrosion inhibitor, or a combination of any two or all three of these additional components. Of various suitable poly- α -olefin oligomers for use in these various additive concentrates, the most preferred are oligomers of 1-decene having a viscosity in the range of 5.5 to 7.5 cSt at 100°C. Also the most preferred detergent/dispersant for such use are the alkenyl succinimides of triethylene tetramine.

The most preferred concentrates of this invention are any of those referred to in the immediately preceding paragraph wherein the components are proportioned such that the concentrate has a density pursuant to ASTM D1298 in the range of 0.9 to 1.0, a flash point pursuant to ASTM D93 in the range of 100 to 120°C, a viscosity at 40°C pursuant to ASTM D445 in the range of 75 to 85 cSt, a pour point pursuant to ASTM D97 below -15°C, and a nitrogen content in the range of 0.55 to 1.20 weight percent.

This invention also provides liquid fuel compositions containing the components of the foregoing additive concentrates of this invention proportioned as above and present in the fuel in an amount at least sufficient to minimize induction system deposits and to control exhaust emissions from an internal combustion engine. While the additives of this invention are suitable for use in diesel fuels, the invention is particularly well adapted for improving performance of gasolines (petrol) especially unleaded gasolines having at least 5 volume % of a fuel-soluble dihydrocarbyl ether and/or an aliphatic monohydric alcohol. Motor gasolines containing at least 10 volume percent of an ether of the formula ROR' where R is methyl or ethyl and R' is tert-butyl or tert-amyl are preferred. Also preferred are fuels wherein the gasoline contains no more than 3% by volume of olefins as determined by ASTM D1319. Most desirably the gasoline contains at least 50% by volume of saturates as determined by ASTM D1319 and contains less than 0.05 weight percent sulfur as determined by ASTM D3120.

Further embodiments involve the use of a plurality of additive components of this invention proportioned as described above in a hydrocarbonaceous fuel for an internal combustion engine (most preferably, gasoline) to decrease total exhaust emissions or fuel consumption, and most preferably both of these.

The foregoing and other advantages and embodiments of this invention will be apparent from the ensuing description and appended claims.

A feature of this invention is that if the particular additive components are not employed, and if employed, are not employed in the proportions specified above, decidedly inferior results can be experienced.

It will be appreciated that while in most cases the practice of this invention will result in reductions in induction system deposits, in total exhaust emissions, and in fuel consumption, in some instances less than all of these advantageous results may be achieved pursuant to this invention.

Component a). Any suitable fuel-soluble succinimide detergent/dispersant can be used, such as for example the succinimides described in Published PCT Patent Application WO 93/06194. However the preferred succinimides are as described in Published European Patent application, Publication No. 0 441 014 (August 14, 1991). For further details, one should refer to that document.

Component b). The carrier oils used in the practice of this invention have viscosities in the range of 10 to 13 cSt at 100°C. The oils can be paraffinic, naphthenic, asphaltic, or blends of two or more such mineral oil base stocks. Preferred mineral oils are those having viscosities in the foregoing range and having the volatility and other characteristics of the mineral oils described in Published British Patent Application, Publication No. 2,259,522 (March 17, 1993). Particularly preferred is a solvent neutral oil falling within the foregoing viscosity range and having a NOACK volatility in the range of 12 to 22% at 300°C.

Component c). Poly- α -olefin oligomers are described in Published European Patent Application, Publication No. 0 526 129 (February 3, 1993) to which reference should be made if details are desired. However in the practice of this invention the oligomer used must have a viscosity in the range of 5 to 10 cSt at 100°C, and preferably the viscosity is in the range of 5.5 to 7.8 cSt at 100°C.

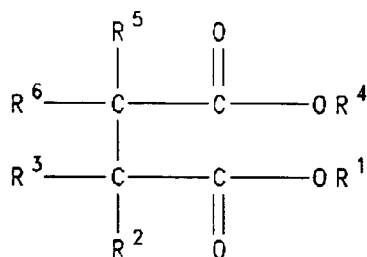
Component d). Aromatic hydrocarbon solvents are available from a number of commercial sources. Small amounts (e.g., up to 5 volume %) of non-aromatic hydrocarbons can be present in the mixture provided they are neither olefinic or acetylenic in character. The important physical properties are the boiling range and viscosity of the mixture, and these properties have been described hereinabove.

Other components which preferably are used include the following:

Antioxidant. Various compounds known for use as oxidation inhibitors can be utilized in the practice of this invention. These include phenolic antioxidants, amine antioxidants, sulfurized phenolic compounds, and organic phosphites, among others. For best results, the antioxidant should be composed predominantly or entirely of either (1) a hindered phenol antioxidant such as 2-tert-butylphenol, 2,6-di-tert-butylphenol, 2,4,6-tri-tert-butylphenol, 4-methyl-2,6-di-tert-butylphenol, 4,4'-methylenebis-(2,6-di-tert-butylphenol), and mixed methylene bridged polyalkyl phenols, or (2) an aromatic amine antioxidant such as the cycloalkyl-di-lower alkyl amines, and phenylenediamines, or a combination of one or more such phenolic antioxidants with one or more such amine antioxidants. Particularly preferred for use in the practice of this invention are tertiary butyl phenols, such as 2,6-di-tert-butylphenol, 2,4,6-tri-tert-butylphenol, and o-tert-butylphenol.

Demulsifier. A wide variety of demulsifiers are available for use in the practice of this invention, including, for example, polyoxyalkylene glycols, oxyalkylated phenolic resins, and like materials. Particularly preferred are mixtures of polyoxyalkylene glycols and oxyalkylated alkylphenolic resins, such as are available commercially from Petrolite Corporation under the TOLAD trademark. One such proprietary product, identified as TOLAD 9308, is understood to be a mixture of these components dissolved in a solvent composed of heavy aromatic naphtha and isopropanol. This product has been found efficacious for use in the compositions of this invention. However, other known demulsifiers can be used such as TOLAD 286.

Corrosion Inhibitor. Here again, a variety of materials are available for use as corrosion inhibitors in the practice of this invention. Thus, use can be made of dimer and trimer acids, such as are produced from tall oil fatty acids, oleic acid, linoleic acid, or the like. Products of this type are currently available from various commercial sources, such as, for example, the dimer and trimer acids sold under the HYSTRENE trademark by the Humko Chemical Division of Witco Chemical Corporation and under the EMPOL trademark by Emery Chemicals. Another useful type of corrosion inhibitor for use in the practice of this invention are the alkenyl succinic acid and alkenyl succinic anhydride corrosion inhibitors such as, for example, tetrapropenylsuccinic acid, tetrapropenylsuccinic anhydride, tetradecenylsuccinic acid, tetradecenylsuccinic anhydride, hexadecenylsuccinic acid, and hexadecenylsuccinic anhydride. Also useful are the half esters of alkenyl succinic acids having 8 to 24 carbon atoms in the alkenyl group with alcohols such as the polyglycols. Preferred materials are the succinic acids or derivatives thereof represented by the formula:



wherein each of R², R³, R⁵ and R⁶ is, independently, a hydrogen atom or a hydrocarbyl group containing 1 to

30 carbon atoms, and wherein each of R¹ and R⁴ is, independently, a hydrogen atom, a hydrocarbyl group containing 1 to 30 carbon atoms, or an acyl group containing from 1 to 30 carbon atoms.

The groups R¹, R², R³, R⁴, R⁵, and R⁶ when in the form of hydrocarbyl groups, can be, for example, alkyl, cycloalkyl or aromatic containing groups. Preferably R¹, R², R³, R⁴ and R⁵ are hydrogen or the same or different straight-chain or branched-chain hydrocarbon radicals containing 1-20 carbon atoms. Most preferably, R¹, R², R³, R⁴, and R⁵ are hydrogen atoms. R⁶ when in the form of a hydrocarbyl group is preferably a straight-chain or branched-chain saturated hydrocarbon radical.

Most preferred is a tetraalkenyl succinic acid of the above formula wherein R¹, R², R³, R⁴ and R⁵ are hydrogen and R⁶ is a tetrapropenyl group.

The following examples in which all parts and percentages are by weight unless otherwise specified, illustrate the practice of this invention. These examples are not intended to limit, do not limit, and should not be construed as limiting the practice of this invention in its generic aspects.

Example 1

An additive concentrate of this invention is prepared by blending together the following components:

240 parts	of polyisobutenyl succinimide detergent/dispersant;
400 parts	of 500 Solvent Neutral mineral oil having a NOACK volatility of 15% at 300°C;
67 parts	of unhydrogenated 1-decene oligomer having a viscosity of 7.3 cSt at 100°C;
240 parts	of an aromatic solvent with a boiling range of 196-256°C and a viscosity of 2.55 cSt at 25°C;
40 parts	of a mixture of 20 parts of N,N'-di-sec-butyl-p-phenylenediamine and 20 parts of a tertiary butylated phenol antioxidant mixture containing a minimum of 75% 2,6-di-tert-butylphenol, 10-15% 2,4,6-tri-tert-butylphenol, and 15-10% of 2-tert-butylphenol; and
13 parts	of TOLAD 286K, demulsifier, available from Petrolite Corporation.

The preferred succinimide dispersant for use in this mixture is formed by reacting polyisobutenyl succinic anhydride with a commercially-available mixture of acyclic and cyclic ethylene polyamines approximating triethylene tetramine in overall composition. The polyisobutenyl succinic anhydride is derived from polyisobutene having a GPC number average molecular weight in the range of approximately 900 to 950. The succinimide is formed in the absence of any diluent or solvent and involves reaction of 1.8 moles of the polyisobutenyl succinic anhydride per mole of triethylene tetramine.

Example 2

A fuel composition of this invention is formed by blending an additive concentrate formed as in Example 1 into an unleaded gasoline to a concentration of 400 parts per million by volume. One preferred gasoline for use in forming this composition contains 12.8 % by volume of methyltertiary-butyl ether and has the following characteristics:

Property	Test Method	Value
Density at 15°C	ASTM D4052	0.772 kg/L
IBP	ASTM D86	42°C
10%	ASTM D86	63°C
50%	ASTM D86	106°C
90%	ASTM D86	154°C
FBP	ASTM D86	199°C
% Off at 70°C	ASTM D86	16 vol %
% Off at 100°C	ASTM D86	45 vol %
% Off at 180°C	ASTM D86	98 vol %
RON	ASTM D2699/86	97.2
MON	ASTM D2700/86	86.0
RVP	ASTM D323	0.49 bar
Sulphur	ASTM D3120	< 0.01%
Aromatics	ASTM D1319	46.9 vol %
Olefins	ASTM D1319	2.4 vol %
Saturates	ASTM D1319	50.8 vol %

Example 3

Three fuels of this invention formed as in Example 2 using the preferred detergent/dispersant of Example 1 and the preferred gasoline of Example 2 were subjected to the Mercedes M102E Inlet Valve Cleanliness Test. The test involved use of the most severe version of the test, namely, CEC F-05-T-92 wherein the inlet valves are pegged to prevent them from rotating. The three fuels of this invention contained respectively 400, 500, and 600 ppm by volume of the additive concentrate. In this example (and in each of the ensuing examples as well), a control test was conducted involving use of the same unleaded base fuel containing the same amount of the methyltertiary-butyl ether but without the additive concentrate of this invention. Table 1 summarizes the test results wherein the higher the merit rating, and the lower the average valve deposit weight, the better. In Table 1 (and in the ensuing tables) "Additive Concentration" refers to the additive concentrate of this invention and thus even though the control fuel always contains the methyltertiary-butyl ether, the term "None" refers to the control fuel.

Table 1

- Mercedes M102E Performance		
Additive Concentration By Volume	Average Valve Merit Rating	Average Deposit Weight, mg/Valve
None	8.16	283
400 ppm	9.14	98
500 ppm	9.30	55
600 ppm	9.40	30

Example 4

As in Example 3, fuels of this invention and the control fuel were subjected to the Renault 5 Test which measures carburetor cleanliness by a merit scale wherein 10 is absolutely clean. The test procedure is defined in CEC-F-03-T-81. Table 2 summarizes the results.

Table 2

<u>- Renault 5 Performance</u>	
Additive Concentration By Volume	Carburetor Merit Rating
None	6.4
350 ppm	7.7
400 ppm	9.1

Example 5

In this instances, a fuel composition formed as in Example 2 was compared to the control fuel in the Peugeot 205 GTi Test developed by the Institute Francais de Petrole to measure fuel injector fouling. This is a 150 hour test that uses a cycle of hot running followed by period of engine shutdown. Table 3 summarizes these test results.

Table 3

<u>- Peugeot 205 GTi Performance</u>	
Additive Concentration By Volume	Average Flow Loss, %
None	1.3
400 ppm	0.1

Example 6

In this pair of tests, the performance of fuel composition of this invention and of the control fuel were compared using the Opel Kadett Inlet Valve Test (CEC F-04-A-87). The results are summarized in Table 4 wherein value 10 in the valve merit rating scale is absolutely clean.

Table 4

<u>- Opel Kadette Inlet Valve Performance</u>		
Additive Concentration By Volume	Average Valve Merit Rating	Average Deposit Weight, mg/Valve
None	5.08	589
400 ppm	8.97	114

Example 7

Two fuels of this invention and the control fuel were subjected to engine tests to evaluate emission performance and fuel consumption. The tests were conducted in a Volkswagen Golf CL vehicle which in each case

had been operated on a particular fuel for deposit accumulation. Then the test fuel was used to operate the vehicle on a chassis dynamometer using the ECE 15-04 cycling procedure during which total emissions were measured. Each test was run two or three times in order to improve the accuracy of the results. Table 5 summarizes the results of these tests.

Table 5

- Emissions Performance and Fuel Consumption					
Additive Concentration By Volume	Total Emissions, g/test				Fuel Consumption, Liters/ 100 km
	HC	NOX	CO	CO ₂	
None	1.633	1.87	10.94	165.5	8.02
400 ppm	1.370	1.96	9.86	163.3	7.82
1000 ppm	1.401	1.96	10.19	163.3	7.84

The results in Table 5 indicate that the compositions of this invention reduced all emissions except NOX, reduced the total of all emissions produced, and improved fuel consumption by 2%.

Example 8

A set of corrosion tests was conducted in which the performance of the fuel of Example 2 was compared to the control fuel using the IP 135 Test. This involves immersing a steel pin in a mix of test fuel and water, and storing for 5 hours at 38°C. The steel pin is then rated according to the NACE corrosion scale in which A denotes no corrosion and E denotes severe corrosion. Both IP 135A and 135B were run in which distilled water and synthetic sea water, respectively, are used. Table 6 summarizes the results.

Table 6

- Corrosion Performance		
Additive Concentration By Volume	IP 135A Rating	IP 135B Rating
None	B	D
400 ppm	A	B

As can be seen from the results of all of the foregoing tests, the practice of this invention gave improved induction system cleanliness, reduced total engine emissions, reduced fuel consumption, and reduced corrosion. It is worth noting that the additive concentrate of this invention decreased corrosion even though the particular composition used did not contain a conventional corrosion inhibitor.

The additive concentrates will normally be employed in amounts providing from 20 to 700, and preferably from 60 to 400 parts by weight of the succinimide detergent/dispersant per million parts by weight of the base fuel. Generally, the additive concentrates of this invention will contain from 10 to 40, and preferably from 20 to 30 weight percent of the succinimide detergent/dispersant based on the total weight of the concentrate.

The fuels of this invention will typically contain from 5 to 80 and preferably from 15 to 40 ppm by weight of antioxidant, from 1 to 30 and preferably from 3 to 15 ppm by weight of demulsifier, and, if used, from 1 to 25 and preferably from 1 to 10 ppm by weight of corrosion inhibitor. In the additive concentrates of this invention, the amounts of these components will be proportioned such that at the recommended dosage level of the concentrate, the fuel will contain the desired amount of each such component within the ranges specified in the immediately preceding sentence. All proportions given herein are in terms of active content of the component. Thus if the detergent/dispersant or any other component as supplied is diluted with a solvent, the amount of the solvent should be excluded from consideration in calculating proportions for use. Care should be exercised in selecting components supplied in liquid diluents or carriers to be sure that such diluents or carriers do not materially detract from or otherwise materially interfere with the performance of the composition.

Numerical ranges of concentrations and proportions given herein are susceptible to minor variations which do not materially alter the performance of the particular composition under consideration.

As used herein, the term "fuel-soluble" means that the material under discussion can be dissolved in the particular fuel under consideration to a concentration at least sufficient to achieve the minimum concentration level specified herein. Preferably the component has a significantly higher solubility in the fuel than such minimum concentration level. However, the term does not mean that the component must be soluble in all proportions in the fuel.

Claims

1. A fuel additive concentrate for minimizing induction system deposits in and controlling exhaust emissions from an internal combustion engine, which concentrate comprises at least the following components:
 - (a) at least one fuel-soluble succinimide detergent/dispersant;
 - (b) a fuel-soluble mineral oil having a viscosity in the range of 10 to 13 cSt at 100°C;
 - (c) unhydrogenated poly- α -olefin oligomer having a viscosity in the range of 5 to 10 cSt at 100°C; and
 - (d) aromatic hydrocarbon boiling in the range of 160 to 300°C and having a viscosity in the range of 1.4 to 2.0 cSt at 25°C;
 wherein the weight ratio of (d):(a) is at least 0.5:1, the weight ratio of (c):(a) is in the range of 0.1:1 to 3:1, the weight ratio of (b):(c) is in the range of 1:1 to 8:1, and wherein the weight ratio of (b)+(c):(a) is in the range of 1.5:1 to 2.5:1.
2. A concentrate according to Claim 1 wherein the weight ratio of (b):(c) is in the range of 3:1 to 7:1; the weight ratio of (d):(a) is in the range of 0.7:1 to 20:1; and the weight ratio of (b)+(c):(a) is in the range of 1.7:1 to 2.2:1.
3. A concentrate according to Claim 1 wherein the weight ratio of (b):(c) is in the range of 5:1 to 7:1; the weight ratio of (d):(a) is in the range of 0.7:1 to 4:1; and the weight ratio of (b)+(c):(a) is in the range of 1.8:1 to 2.1:1.
4. A concentrate according to Claim 1 wherein the weight ratio of (b):(c) is in the range of 5.5:1 to 6.5:1; the weight ratio of (d):(a) is in the range of 0.9:1 to 1.5:1; and the weight ratio of (b)+(c):(a) is in the range of 1.8:1 to 2.1:1.
5. A concentrate according to any of the preceding claims wherein the unhydrogenated poly- α -olefin oligomer is an oligomer of 1-decene having a viscosity in the range of 5.5 to 7.5 cSt at 100°C.
6. A concentrate according to any of the preceding claims further comprising at least one fuel-soluble anti-oxidant, or at least one fuel-soluble demulsifying agent, or at least one fuel-soluble corrosion inhibitor, or a combination of any two or all three of these additional components.
7. A concentrate according to any of the preceding claims wherein the detergent/dispersant is an alkenyl succinimide of triethylene tetramine.
8. A concentrate in accordance with any of the preceding claims further characterized in that said concentrate has a density pursuant to ASTM D1298 in the range of 0.9 to 1.0, a flash point pursuant to ASTM D93 in the range of 100 to 120°C, a viscosity at 40°C pursuant to ASTM D445 in the range of 75 to 85 cSt, a pour point pursuant to ASTM D97 below -15°C, and a nitrogen content in the range of 0.65 to 1.20 weight percent.
9. A liquid fuel composition which comprises a hydrocarbonaceous fuel containing the components in accordance with any of Claims 1-8 in an amount at least sufficient to minimize induction system deposits and to control exhaust emissions from an internal combustion engine.
10. A fuel composition in accordance with Claim 9 wherein the hydrocarbonaceous fuel is gasoline.
11. A fuel composition in accordance with Claim 9 or Claim 10 wherein the composition further comprises at least 5 volume percent of at least one fuel-soluble dihydrocarbyl ether or aliphatic monohydric alcohol.
12. A fuel composition in accordance with Claim 9 wherein the fuel is gasoline containing at least 10 volume percent of an ether of the formula ROR' where R is methyl or ethyl and R' is tert-butyl or tert-amyl.

13. A fuel composition in accordance with Claim 12 wherein the gasoline contains no more than 3% by volume of olefins as determined by ASTM D1319.
- 5 14. A fuel composition in accordance with Claim 12 or 13 wherein the gasoline contains at least 50% by volume of saturates as determined by ASTM D1319 and contains less than 0.05 weight percent sulfur as determined by ASTM D3120.
- 10 15. The use of a plurality of additive components in accordance with any one of Claims 1-8 in a hydrocarbonaceous fuel for an internal combustion engine to decrease total exhaust emissions and/or fuel consumption.
- 15 16. The use of an additive concentrate in accordance with any one of Claims 1-8 in gasoline in an amount at least sufficient to decrease total exhaust emissions and/or fuel consumption of an internal combustion engine operated on the resultant fuel composition.
17. A use in accordance with Claim 16 wherein the resultant fuel composition is in accordance with any of Claims 12-14.

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European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 94 30 5054

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
D,X	EP-A-0 526 129 (ETHYL PETROLEUM ADDITIVES)	I,2,5-7, 9-12	C10L1/14
Y	* the whole document * ---	15-17	C10L10/00
Y	DATABASE WPI Week 9329, Derwent Publications Ltd., London, GB; AN 93-232635 & JP-A-5 156 270 (TONEN) * abstract * ---	15-17	C10L10/02
P,X	EP-A-0 569 228 (ETHYL PETROLEUM ADDITIVES) * the whole document * ---	1,2,5-7, 9-14	
D,X	EP-A-0 441 014 (ETHYL PETROLEUM ADDITIVES) * page 3, line 37 - line 54; claims 1-10; example 1 * * page 4 * * page 5, line 10 - line 21 * ---	1,2,6,7, 9,10	
X	GB-A-1 486 144 (CITIES SERVICE OIL COMP.) * the whole document * ---	1,2,6,9, 10	
A	US-A-5 006 130 (AIELLO ET AL.) * the whole document * ---	1-17	
A	WO-A-92 02601 (MOBIL OIL CORP.) * page 13 - page 14; claims 1-24 * ---	1-17	
A	EP-A-0 476 196 (ETHYL PETROLEUM ADDITIVES) * the whole document * -----	15-17	
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 20 September 1994	Examiner De La Morinerie, B
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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