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(71) Applicant: **NIPPON MINING COMPANY LIMITED**
12-32, Akasaka 1-chome Minato-ku
Tokyo(JP)

Applicant: **KYODO OIL TECHNICAL RESEARCH CENTER CO., LTD.**
12-32 Akasaka 1-chome
Minato-ku Tokyo(JP)

(72) Inventor: **Date, Kazuto c/o Kyodo Oil Technical Research**
Center Co.,Ltd 17-35,Niizominami 3-chome
Toda-shi, Saitama-ken(JP)
Inventor: **Tamanouchi, Mitsuo c/o Kyodo Oil Technical**
Center Co.,Ltd 17-35,Niizominami 3-chome
Toda-shi, Saitama-ken(JP)
Inventor: **Kudoh, Masuo c/o Kyodo Oil Technical Center**
Co.,Ltd 17-35,Niizominami 3-chome
Toda-shi, Saitama-ken(JP)
Inventor: **Tanaka, Hideo c/o Technical Research Center,**
Nippon Mining Co.,Ltd 17-35,Niizominami 3 ch.
Toda-shi, Saitama-ken(JP)
Inventor: **Kawate, Akihiro**
c/o Mizushima Oil Refinery,Nippon Mining Co.,Ltd
2-1,Ushiodori,Kurashiki,Okayama-ken(JP)
Inventor: **Akada, Takumi**
c/o Mizushima Oil Refinery,Nippon Mining Co.,Ltd
2-1, Ushiodori,Kurashiki-shi,Okayama-ken(JP)
Inventor: **Sasaki, Takanobu**
c/o Nippon Mining Co., Ltd. 12-32, Akasaka 1-chome
Minato-ku Tokyo(JP)

(74) Representative: **Harrison, David Christopher et al**
MEWBURN ELLIS & CO 2/3 Cursitor Street
London EC4A 1BQ(GB)

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㊹ Cetane number improver additive for diesel engine fuels, fuelcomposition and fuel production process.

㊼ Disclosed is a novel cetane number improver additive for diesel engine fuels comprising tetralin hydroperoxide as the active ingredient. A diesel engine fuel composition having a high cetane number may be obtained by adding this additive to a diesel engine fuel in such an amount as to adjust its peroxide number to a value of 100 to 1,000. Also disclosed is a process for the production of a diesel engine fuel which comprises the steps of hdyrogenating a naphthalene- or alkylnaphthalenes-containing hydrocarbon oil and partially oxidizing the hydrogenated oil to yield a hydrocarbon oil containing tetralin hydroperoxide.

Cetane Number Improver Additive for Diesel Engine Fuels, Fuel Composition and Fuel Production Process

Background of the Invention

1. Field of the Invention

This invention relates to a cetane number improver additive for diesel engine fuels, a diesel engine fuel composition containing the additive, and a process for the production of a diesel engine fuel having a high cetane number. Furthermore, it also relates to a technique for utilizing cycle oil obtained by fluidized catalytic cracking of a heavy hydrocarbon oil, tar oil obtained as a by-product by cracking of naphtha, and oil obtained by liquefaction of coal, as diesel engine fuel components.

2. Description of the Prior Art

If diesel engine fuels (i.e., gas oil and fuel oil) do not have an adequately high cetane number, the so-called diesel knock phenomenon will take place. This may produce loud engine noises and cause a reduction in combustion efficiency. If the cetane number becomes still lower, it may be impossible to start the engine.

In order to fully utilize light cycle oil obtained by fluidized catalytic cracking of a heavy hydrocarbon oil and having a high aromatics content (hereinafter referred to as FCC-LCO), oil obtained by liquefaction of coal, and tar oil obtained as a by-product during the production of ethylene by cracking of naphtha, attempts have recently been made to hydrogenate such oils and use the resulting oils as diesel engine fuel components. However, these oils have a very low cetane number of the order of 15 to 25 and contain a considerable amount of polycyclic aromatic compounds (mainly comprising naphthalene and its derivatives), so that it is impossible to mix a high proportion of these oils with other straight-run petroleum oils (i.e., straight-run fractions obtained by distillation of petroleum, such as gas oil and the like).

Conventionally, a large number of additives for improving the cetane number of diesel engine fuels have been proposed and some of them are now on the market.

Among these cetane number improver additives are nitrate compounds such as tetrahydro-2,5-furan dimethanol nitrate (U.S. Patent 4,522,630), alkyl and alkoxy nitrates (U.S. Patents 4,549,883 and 4,448,587), cyclododecyl nitrate (U.S. Patent 4,420,311), polysaccharide nitrate esters (Japanese Patent Laid-Open No. 25387/'83) and the like. However, these additives are disadvantageous from an economical point of view, because the desired effect cannot be obtained unless they are added in an amount of as much as 1,000 to 3,000 ppm.

In addition to the above-described additives, other techniques for improving combustibility have also been proposed. For example, OLS 3136030 discloses a method in which dialkoxyalkanes are partially oxidized with ozone or H_2O_2, and U.S. Patent 4,330,304 discloses a combustion improver additive comprising nitroparaffin, cumene hydroperoxide and propylene oxide. Furthermore, the present inventors have previously found that the ignitability of a diesel engine fuel can be improved by oxidizing the fuel partially and then feeding it to a diesel engine (Japanese Patent Laid-Open Nos. 45765/'85, 233703/'83 and 24386/'84) and that cumene hydroperoxide can act as a cetane number improver additive (Japanese Patent Laid-Open No. 27767/'85).

Diesel engines have wide applications including vehicles (such as trucks, buses, passenger cars, locomotives, etc.), construction machinery, electric power generators, ships and the like. In these diesel engines, gas oil and fuel oil are used as fuels. Specifically, the fuels for high-speed and small-sized diesel engines used in trucks, buses and the like have conventionally been produced chiefly from an oil obtained by hydrogenating a petroleum distillate having a boiling range of about 230 to about 400° C. On the other hand, the heavy fuels for medium- to large-sized diesel engines used in ships and the like have been produced chiefly from an oil obtained by hydrogenating reduced crude. These fuels have a cetane number of about 45 to about 60 and are suitable for use in diesel engines. However, crackers for heavy hydrocarbon oils and naphthas are now being widely used in petroleum refining industry and petrochemical industry, so that cracked oils and tar oils containing a high proportion of aromatics (such as naphthalene and the like) and having a low cetane number are being produced in large amounts. In addition, the development of coal liquefaction equipment has led to the production of fuel oils having a high aromatics content and a low cetane number. In order to use these oils as diesel engine fuels, a technique for improving their cetane number is required. The present inventors have made an intensive study in the above-described background and have completed this invention.

Summary of the Invention

The present invention has been completed on the basis of the discovery that tetralin hydroperoxide, which is a partial oxidation product of tetralin, is effective in improving the cetane number of diesel engine fuels. When cetane numbers were measured using a cetane number measuring engine (CFR engine), the present inventors noticed that the performance of tetralin used as the standard fuel varied from lot to lot. As a result of careful search for its causes, it was found that the tetralin contained a slight amount of tetralin hydroperoxide and its treatment with clay gave a stabilized value. This has led to the discovery that tetralin hydroperoxide can be effectively used as a cetane number improver additive. On the basis of this discovery, the present inventors have investigated the method of forming tetralin hydroperoxide in a fuel production process and have thereby established a process for the production of a fuel having a high cetane number from a fuel material having a low cetane number.

Accordingly, it is an object of the present invention to provide a novel cetane number improver additive for diesel engine fuels comprising tetralin hydroperoxide as the active ingredient.

It is another object of the present invention to provide a diesel engine fuel composition having a high cetane number.

It is still another object of the present invention to provide a process for producing a diesel engine fuel having a high cetane number from a fuel material having a low cetane number.

In order to accomplish these objects, the present invention provides:

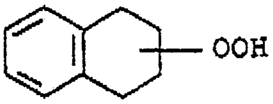
(1) a cetane number improver additive for diesel engine fuels comprising tetralin hydroperoxide as the active ingredient;

(2) a diesel engine fuel composition comprising (a) a diesel engine fuel and (b) the aforesaid additive which is added to the diesel engine fuel in such an amount as to adjust its peroxide number to a value of 100 to 1,000; and

(3) a process for the production of a diesel engine fuel which comprises the steps of (a) hydrogenating a naphthalene- or alkylnaphthalenes-containing hydrocarbon oil to effect partial nuclear hydrogenation and thereby convert at least a part of the naphthalene or alkylnaphthalenes into tetralin, and (b) partially oxidizing the hydrogenated oil to convert the tetralin into tetralin hydroperoxide.

Detailed Description of the Invention

The cetane number improver additive for the present invention comprises, as the active ingredient, tetralin hydroperoxide having a structure represented by the formula



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This compound can be readily formed by exposing tetralin to ultraviolet light in air, or by heating tetralin in an atmosphere of oxygen (preferably in the presence of copper wire). For example, when tetralin is placed in an atmosphere of pure oxygen having a gauge pressure of 3 to 10 kg/cm^2 and allowed to stand at a temperature of 8 to 100 °C for about 6 hours or so, a part of the tetralin is oxidized to form tetralin hydroperoxide in such an amount as to give a peroxide number of about 1,000 to 2,000. Peroxide number can be measured according to the procedure described in ASTM D1563-84, "Standard Test Method for Peroxide Number of Mineral Insulating Oils". In order to use the resulting oil as an additive, it is convenient for blending purposes to dilute the oil with an n-paraffin having 6 to 16 carbon atoms. In this connection, the cetane number improving effect of peroxides is reported in SAE Quarterly Transactions, Vol. 5, No. 3, pp. 404-417 (1951), but no mention of tetralin hydroperoxide is found therein. This publication discloses that dibutyl peroxide, when added to a diesel engine fuel in an amount of 1.5% by volume, increases its cetane number by 20. In contrast, the additive of the present invention exhibits a marked effect at considerably lower concentrations, has good storage stability, and is easy to handle. More specifically, the additive of the present invention may be added to a diesel engine fuel in such an amount as to adjust its peroxide number to a value of about 100 to 1,000, thus increasing its cetane number by about 3 to 15.

The present invention also relates to a process for the production of a diesel engine oil which comprises the steps of hydrogenating a naphthalene-or alkylnaphthalenes-containing hydrocarbon oil to convert at least of the naphthalene or alkylnaphthalenes into tetralin or alkyl derivatives thereof, and partially oxidizing the hydrogenated oil to yield a hydrocarbon oil containing tetralin hydroperoxide.

The raw oil used in the present invention is a hydrocarbon oil containing a considerable amount of naphthalene or alkylnaphthalenes. Specific examples thereof include light cycle oil obtained by fluidized catalytic cracking of a heavy hydrocarbon oil (i.e., FCC-LCO), oil obtained by liquefaction of coal, and tar oil obtained as a by-product by cracking of naphtha.

FCC-LCO useful as a raw oil has a boiling range of about 150 to 350°C and is characterized by n-d-M ring analysis values of 45-65% C_A, 0.5-5% C_N and 35-50% C_p, an aniline point of 20 to 30, and a cetane number of about 10 to 20.

Useful oil obtained by liquefaction of coal is a gas oil-to-fuel oil A fraction having a boiling range of about 150 to 450°C. This oil is recovered from coal liquefaction equipment and has a cetane number of 15 to 20 and an aromatics content of about 60% by weight. Useful tar oil obtained as a by-product by cracking of naphtha is a fraction having a boiling range of about 150 to 400°C and containing a high proportion of aromatics. In these raw oils, naphthalene or alkylnapthalens having one or more alkyl substituents of 1 to 5 carbon atoms are present in an amount of about 4 to 20% by weight.

In the process of the present invention, these raw oils are subjected to hydrogenation and partial oxidation under mild conditions.

The hydrogenation is carried out in the presence of a hydrogenation catalyst commonly used in petroleum refining. Specific examples thereof are catalysts formed by supporting one or more metals (such as Ni, Co, Mo, W, V, Fe and the like) on a suitable carrier (such as alumina, silica, silica-alumina and the like). In particular, it is preferable to use a catalyst and reaction conditions suitable for partial nuclear hydrogenation of naphthalene and derivatives thereof, i.e., suitable for their conversion into tetralin and/or alkyl derivatives thereof. Among others, W-Ni catalysts are especially preferred for nuclear hydrogenation of naphthalene. Although the hydrogenation conditions can vary according to the types of raw oil and catalyst used, the hydrogenation is usually carried out in the presence of an Ni-W catalyst and under conditions including a reaction pressure (hydrogen pressure) of 40 to 120 kg/cm^2, a temperature of 250 to 350°C and preferably 300 to 340°C, an LHSV of 0.4 to 4.0 hr^{-1} and preferably 1.5 to 2.0 hr^{-1}, and a hydrogen feed rate of 300 to 1,000 ℓ/ℓ of the raw oil.

The hydrogenation conditions should be selected so that a reaction can take place in which the naphthalene or alkylnaphthalenes present in the raw oil undergo partial nuclear hydrogenation and form tetralin and/or alkyl derivatives thereof. Although the hydrogenation conditions can vary according to the type of catalyst used, they should be selected with due consideration to the criterion that the content of tetralin and/or alkyl derivatives thereof present in the hydrogenated oil should be as high as possible, i.e., not less than 0.5% by weight, preferably not less than 1.0% by weight and more preferably not less than 1.5% by weight. The tetralin and/or alkyl derivatives thereof are formed by partial nuclear hydrogenation of the naphthalene or alkylnaphthalenes. Accordingly, no upper limit is placed on the content of such naphthalene compounds in the raw oil, and higher contents are more effective. However, a content of about 3 to about 16% by weight will suffice. In the aforesaid FCC-LCO and oil obtained by liquefaction of coal, such naphthalene compounds are present in an amount of not less than 4% by weight.

As used herein, the term "alkyl derivatives of tetralin" means derivatives of tetralin having 1 to 3 alkyl substituents each containing 1 to 5 carbon atoms.

The hydrogenation conditions which cause complete nuclear hydrogenation of the naphthalene or alkylnaphthalenes present in the raw oil (i.e., those which convert the naphthalene or alkylnaphthalenes into decalin or derivatives thereof) should be avoided. For this reason, severe hydrogenation conditions should be avoided because they convert the naphthalene into decalin.

Accordingly, the hydrogenation conditions, such as type of catalyst, reaction temperature, liquid space velocity and the like, should be selected so that the hydrogenated oil will contain decelin or derivatives thereof in an amount of not greater than 5% by weight, preferably not greater than 3% by weight and more preferably not greater than 1% by weight and, at the same time, will contain tetralin or derivatives thereof in an amount of not less than 0.5% by weight and preferably not less than 1.0% by weight. The formation of a large amount of decalin or derivatives thereof causes a decrease in the content of tetralin or derivatives thereof and, therefore, is undesirable for subsequent partial oxidation.

Another criterion useful in determining the hydrogenation conditions is that the n-d-M ring analysis values of the hydrogenated oil should be 20-35% C_A, 30-45% C_N and 30-35% C_P, and preferably 25-35% C_A and 30-40% C_N.

Oil obtained by liquefaction of coal and tar oil obtained by cracking of naphtha contain a large amount of naphthalene compounds. These oils should likewise be hydrogenated under such conditions that the naphthalene compounds are selectively converted into tetralin or derivatives thereof and their conversion of into decalin or derivatives thereof is suppressed. At the hydrogenation conditions become severer, the hydrogenated oil has a higher cetane number. However, it is desirable that the cetane number of the hydrogenated oil be limited to at most 45 or so.

The oil obtained by the above-described hydrogenation is then subjected to partial oxidation. The term "partial oxidation" as used herein means a process for increasing the peroxide number of the oil to a value of not less than 100. The peroxide number of the oil can be measured according to the procedure described in ASTM D1563-84. The partial oxidation may be carried out by placing the oil under an oxygen pressure of 3 to 8 kg/cm^2 and allowing it to stand at a temperature of 60 to 100 °C for a period of 3 to 10 hours, or by adding a copper of nickel catalyst to the oil and stirring it under milder conditions. However, no particular limitation is placed on the partial oxidation conditions. Thus, the peroxide number of the oil can be increased to a value of not less than 100, preferably not less than 150 and more preferably not less than 300. As the peroxide number becomes higher, the oil has a higher cetane number. In particular, the use of an oil having a high tetralin content provides a peroxide number of not less than 1,500, resulting in a fuel having a high cetane number. More specifically, as a result of the partial oxidation, the tetralin or derivatives thereof present in the oil are readily converted into tetralin hydroperoxide, which functions to markedly improve the cetane number of the oil. The partial oxidation may cause the oil to assume a yellow to brown color, but this color may be improved by treatment with clay or the like. Prior to the partial oxidation, the tetralin content of the hydrogenated oil may further increased by adding a small amount of tetralin. This serves to further improve the cetane number of the partially oxidized oil. It is to be understood in this connection that, if the hydrogenation is carried out under severe conditions which convert naphthalene into deccalin, subsequent partial oxidation will fail to bring about an improvement in cetane number.

The above-described treatment causes the cetane number of the resulting fuel oil to be increased by 30 to 60, as compared with that of the starting oil. For example, when FCC-LCO having a cetane number of 14 is used as the starting oil, its hydrogenation and partial oxidation makes it possible to produce a fuel oil having a cetane number of 50 to 75. This fuel oil may be used directly as a diesel engine fuel or mixed with other fuel components to yield a diesel engine fuel.

The present invention and its effects will be more specifically explained with reference to the following examples.

Example 1

Light cycle oil obtained by fluidized catalytic cracking of a heavy hydrocarbon oil and having the properties shown in Table 1 (i.e., FCC-LCO) was used as the starting oil. According to the following procedure, this starting oil was hydrogenated and then partially oxidized.

In the hydrogenation step, a W-Ni catalyst was used, the reaction pressure (hydrogen pressure) was 100 kg/cm^2, and the temperature and liquid space velocity (LHSV) were as shown in Table 1.

As a result of this hydrogenation, there were obtained hydrogenated oils H-1, H-2 and H-3 as shown in Table 1.

A glass container was placed in a pressure vessel and charged with 50 cc of each of the hydrogenated oils. Then, the pressure vessel was filled with pure oxygen to a pressure of 7.0 kg/cm^2, placed in a thermostatic chamber kept at 100 °C, and allowed to stand for 8 hours. Thereafter, the pressure vessel was allowed to cool and taken out. In this manner, 100 cc of a partially oxidized oil was obtained from each of the hydrogenated oils. The peroxide number and cetane number of the partially oxidized oil obtained from each of the hydrogenated oils were measured and the results thus obtained are shown in Table 2.

Cetane number measurements were made in the following manner: 100 cc of each of the partially oxidized oils was mixed with 900 cc of a commercially available gas oil (having a cetane number of 48.4). Using a CFR engine, the cetane number of this mixed oil was measured according to the procedure described in ASTM D613. In Table 2, the measured cetane numbers of the mixed oils are given in the upper row and the estimated cetane numbers of the partially oxidized oils in the lower row.

Peroxide number measurements were made according to the procedure described in ASTM D1563-84.

Table 1

		Starting oil FCC-LCO	Hydrogenated oil		
			H-1	H-2	H-3
Specific gravity		0.9197	0.8948	0.8770	0.8530
Sulfur content (wt%)		0.29	0.05	0.01	0.01 or less
n-d-M ring analysis	% C_A	55.2	32.7	26.1	15.7
	% C_N	2.6	35.1	42.7	51.1
	% C_P	42.2	32.2	31.2	33.2
$\overline{MW}$		174	170	163	158
Cetane number		14	17	36	47
Tetralin (wt%)		0	1.44	0.98	0.33
Decalin and its derivatives (wt%)		0	0.45	2.98	6.40
Naphthalene (wt%)		4.1	0.16	0.27	0.20
Hydrogenation conditions	Temperature (°C)	-	320	350	380
	LHSV (hr^{-1})	-	2.0	1.5	0.8

Table 2

		Partially oxidized oil from H-1	Partially oxidized oil from H-2	Partially oxidized oil from H-3
Peroxide number		2650	1250	8
Cetane number	Measured value of mixed oil	50.8	50.1	48.2
	Estimated value of partially oxidized oil	72	65	46

It is evident from Table 2 that, when the starting oil FCC-LCO having a cetane number of 14 as shown in Table 1 was hydrogenated and partially oxidized under appropriate conditions, the resulting products (i.e., the partially oxidized oils from H-1 and H-2) exhibited an increased cetane number of 60-70. In contrast, when the starting oil FCC-LCO was hydrogenated under severe conditions and then partially oxidized, the resulting product (i.e., the partially oxidized oil from H-3) showed no appreciable improvement in cetane number. Thus, the hydrogenation should be carried out under such conditions that the content of tetralin is increased and the formation of decalin is suppressed, and this enhances the effect of improving the cetane number by subsequent partial oxidation.

Example 2

(1) Preparation of tetralin hydroperoxide

A glass container having a capacity of 200 cc was placed in a pressure vessel made of stainless steel. After the glass container was charged with 100 cc of tetralin (reagent grade), oxygen gas was introduced into the pressure vessel until an oxygen pressure of 70 kg/cm^2 (G) was reached. Then, the pressure vessel having the glass container placed therein was immersed in a water bath at 100° C and allowed to stand for 6 hours. As a result, a part of the tetralin was partially oxidized to form tetralin hydroperoxide. Thus, a tetralin hydroperoxide-containing tetralin solution having a peroxide number of 1,550 was recovered from the glass container within the pressure vessel.

Separately, the same procedure as described above was repeated except that the time of immersion was increased from 6 hours to 10 hours. Thus, there was obtained a tetralin hydroperoxide-containing tetralin solution having a peroxide number of 2,600.

(2) Preparation of fuel compositions

0.1 liter of each of the above tetralin hydroperoxide-containing tetralin solutions having peroxide numbers of 1,550 and 2,600 was added to 0.9 liter of a fuel having the composition given below. Thus, there were obtained tetralin hydroperoxide-containing fuel compositions having peroxide numbers of 155 and 260, respectively.

(Composition of fuel)

Straight-run gas oil (DS-GO)	60 vol%
Light cycle oil obtained by fluidized catalytic cracking (FC-LCO)	40 vol%

(Cetane number measurement of fuel compositions)

Using a CFR engine (manufactured by Waukesha Co.), the cetane numbers of the above fuel compositions were measured according to the procedure described in ASTM D613.

For purposes of comparison, further fuel compositions were prepared by adding the same amount of tetralin, instead of the tetralin hydroperoxide-containing tetralin solution, to the aforesaid fuel, or by adding commercially available cumene hydroperoxide (reagent grade) in such an amount as to give a peroxide number of 150 or 260. The cetane numbers of these fuel compositions were also measured in the same manner as described above. The results thus obtained are shown in Table 3.

Table 3

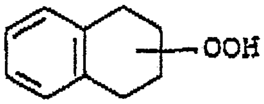
	Base fuel	Additive (amount as peroxide number)	Cetane number
Control	DS-GO 54 vol% FL-LCO 36 vol% Tetralin 10 vol%	None	38.7
Present invention	Ditto	Tetralin hydro-peroxide (155)	42.3
	Ditto	Tetralin hydro-peroxide (260)	44.0
Prior art	Ditto	Cumene hydro-peroxide (150)	40.6
	Ditto	Cumene hydro-peroxide (260)	41.8

As can be seen from Table 3, the cetane numbers of the fuel compositions containing tetralin hydroperoxide were increased by about 3 to 5, as compared with that of the fuel composition containing no additive.

As described above, the fuel composition of the present invention may be produced by using, as the starting material, an oil containing a high proportion of aromatic hydrocarbons and hence having poor combustibility, and modifying it through a very simple means. Thus, the present invention makes it possible to widen the application of materials which have hitherto been unsuitable for use in diesel engine fuels. Furthermore, the cetane number improver additive of the present invention may be readily prepared, is easy to handle, and can exhibit a marked effect.

Claims

1. A cetane number improver additive for diesel engine fuels comprising, as the active ingredient, tetralin hydroperoxide of the formula



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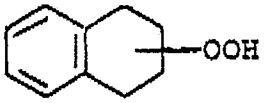
2. A diesel engine fuel composition comprising (a) a diesel engine fuel and (b) tetralin hydroperoxide of the formula



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which is added to the diesel engine fuel in such an amount as to adjust its peroxide number to a value of 100 to 1,000.

3. A diesel engine fuel composition as claimed in claim 2 wherein, before the addition of tetralin hydroperoxide, the diesel engine fuel has a cetane number of not greater than 40.

4. A diesel engine fuel composition comprising (a) a diesel engine fuel and (b) an oil obtained by partially oxidizing tetralin or a tetralin-containing hydrocarbon oil to convert the tetralin into tetralin hydroperoxide, and added to the diesel engine fuel in such an amount as to adjust its peroxide number to a value of 100 to 1,000.

5. A process for the production of a diesel engine fuel which comprises the steps of (a) hydrogenating a naphthalene- or alkylnaphthalenes-containing hydrocarbon oil in the presence of a hydrogenation catalyst to convert at least a part of the naphthalene or alkylnaphthalenes into tetralin and then (b) partially oxidizing the hydrogenated oil to yield a hydrocarbon oil containing tetralin hydroperoxide.

6. A process as claimed in claim 5 wherein the naphthalene- or alkylnaphthalenes-containing hydrocarbon oil comprises one or more oils selected from the group consisting of cycle oil obtained by fluidized catalytic cracking of a heavy hydrocarbon oil derived from petroleum, a diesel fuel fraction obtained by liquefaction of coal, and tar oil obtained by cracking of naphtha.

7. A process as claimed in claim 5 wherein the hydrogenation step is carried out under such conditions that the resulting oil has a tetralin content of not less than 0.5% by weight.

8. A process as claimed in claim 5 wherein the partial oxidation step is carried out under such conditions that the resulting oil has a peroxide number of not less than 100.

9. A process as claimed in claim 5 wherein the partial oxidation step is carried out under such conditions that the resulting oil has a peroxide number of not less than 150.

10. A process for the production of a diesel engine fuel which comprises mixing the diesel engine fuel obtained by the process of claim 5, with another diesel engine fuel.

European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 88 30 2532

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.4)
X,P	US-A-4 723 963 (TAYLOR) * Abstract; claims; column 4, lines 25-68; columns 16-20 * ---	1-10	C 10 L 1/18
X	US-A-2 011 297 (MOSER et al.) * Whole document * ---	1-10	
X	DE-C- 617 492 (BATAAFSCHE PETROLEUM MAATSCHAPPIJ) * Whole document * ---	1-3	
X	DE-C- 651 771 (DR. H. HOCK) * Whole document * ---	1-3	
A	US-A-2 430 864 (FARKAS et al.) * Whole document * ---	1-10	
A	US-A-2 430 865 (FARKAS et al.) * Whole document * -----	1-10	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 10 L

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	19-07-1988	DE LA MORINERIE B.M.S.B.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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