⑲ Europäisches Patentamt
European Patent Office
Office européen des brevets

⑪ Publication number: **0 328 642 A1**

⑫ EUROPEAN PATENT APPLICATION published in accordance with Art. 158(3) EPC

㉑ Application number: **87904959.1**

㉒ Date of filing: **30.07.87**

(86) International application number:
PCT/JP87/00565

(87) International publication number:
WO 89/01021 (09.02.89 89/04)

(51) Int. Cl.⁴: **C10M 111/04 , //C10N40:04**

(43) Date of publication of application:
23.08.89 Bulletin 89/34

(84) Designated Contracting States:
DE FR GB

(71) Applicant: **TOA NENRYO KOGYO KABUSHIKI KAISHA**
1-1 Hitotsubashi, 1-Chome Chiyoda-Ku
Tokyo 100(JP)

(72) Inventor: **YOSHIMURA, Narihiko**
4-3-326, Nishitsurugaoka 1-chome
Ooi-machi, Iruma-gun Saitama 354(JP)
Inventor: **TOMIZAWA, Hirotaka**
29-12, Nakaarai 5-chome Tokorozawa-shi
Saitama 359(JP)
Inventor: **KOMATSU, Yasuji**
1098-7, Kamekubo Ooi-machi, Iruma-gun
Saitama 354(JP)

(74) Representative: **Holmes, Michael John et al**
Frank B. Dehn & Co. Imperial House 15-19
Kingsway
London WC2B 6UZ(GB)

(54) TRACTION FLUID.

(57) A traction fluid which is prepared by compounding 0.1 to 95 wt % of a branched poly-α-olefin in a diester represented by general formula (I) (wherein A' represents an ester bond of -COO- or -OOC-, n represents 1 to 10, R_1 groups represent one or two members selected from among a hydrogen atom and an alkyl group having 1 to 8 carbon atoms, and R_2 groups represent one or two members selected from among a hydrogen atom and an alkyl group having 1 to 3 carbon atoms) or its derivative is disclosed. This fluid is preferably used in a power transmission unit, paricularly a traction drive unit.

TRACTION FLUID

FIELD OF THE INVENTION

This invention relates to a traction fluid. More particularly, the present invention is concerned with a traction fluid comprising a diester or its derivative having two cyclohexyl rings, and a branched poly-α-olefin as the base oil.

BACKGROUND OF THE INVENTION

Traction drive power transmissions, which transmit power to a driven part through a traction drive mechanism, have attracted attention in the field of automobiles and industrial machinery, and in recent years research and development thereon has progressed. The traction drive mechansim is a power transmitting mechanism using a rolling friction. Unlike conventional drive mechanisms it does not use any gears, which enables a reduction in vibration and noise as well as a smooth speed change in high-speed rotation. An important goal in the automobile industry is improvement in the fuel economy of automobiles. It has been suggested that if the traction drive is applied to the transmission of automobiles to convert the transmission to a continuous variable-speed transmission the fuel consumption can be reduced by 20% or more compared to conventional transmission systems since the drive can always be in the optimum speed ratio. Recent studies have resulted in the development of materials having high fatigue resistance and in the theoretical analysis of traction mechanisms. Regarding the traction fluid, the correlation of traction coefficients is gradually being understood on a level of the molecular structure of the components. The term "traction coefficient" as used herein is defined as the ratio of the tractional force which is caused by slipping at the contact points between rotators which are in contact with each other in a power transmission of the rolling friction type to the normal load.

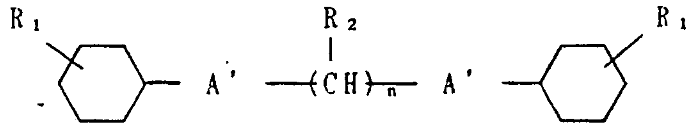
The traction fluid is required to be comprised of a lubricating oil having a high traction coefficient. It has been confirmed that a traction fluid possessing a molecular structure having a naphthene ring exhibits a high performance. "Santotrack ®" manufactured by the Monsanto Chemical Company is widely known as a commercially available traction fluid. Japanese Patent Publication No. 35763/1972 discloses di(cyclohexyl)-alkane and dicyclohexane as traction fluids having a naphthene ring. This patent publication discloses that a fluid obtained by blending the above-mentioned alkane compound with a perhydrogenated (α -methyl)-styrene polymer, hydrindane compound or the like has a high traction coefficient. Further, Japanese Patent Early-publication No. 191797/1984 discloses a traction fluid containing an ester compound having a naphthene ring. It teaches that an ester obtained by the hydrogenation of the aromatic nucleus of dicyclohexyl cyclohexanedicarboxylate or dicyclohexyl phthalate is preferable as the traction fluid.

As mentioned above, there has been progress in recent years in the development of continuous variable-speed transmissions. The higher the traction coefficient of the traction fluid the larger the transmission force in the device. This contributes to a reduction in the size of the device with a corresponding reduction in exhaust gas, thereby reducing environmental pollution. Therefore, there is a demand for a fluid having a traction coefficient as high as possible. However, even the use of a traction fluid which exhibits the highest performance of all the currently commercially available fluids in such a traction drive device provides unsatisfactory performance with respect to the traction coefficient and economics. The traction fluid which has been proposed in Japanese Patent Publication No. 35763/1971 contains Santotrack ® or the like as one component, which is unsatisfactory with respect to performance and economics.

DETAILED DESCRIPTION OF THE INVENTION

The present inventors have made extensive and intensive studies with a view to developing a traction fluid which not only exhibits a high traction coefficient but which also is low in cost. As a result, they have found that the incorporation of a specified amount of branched poly-α-olefin into a diester or its derivative in which two cyclohexyl rings are connected through a linear chain hydrocarbon can provide an economical, high-performance base oil fluid, and achieved the present invention.

The present invention relates to a traction fluid characterized by being a blend of a diester or its derivative represented by the following general formula



wherein A′ is an ester linkage of -COO- or -OOC-, n is an integer of 1 to 10, R_1 is independently selected from a hydrogen atom and alkyl groups having 1 to 8 carbon atoms, and R_2 is independently selected from a hydrogen atom and alkyl groups having 1 to 3 carbon atoms: with 0.1 to 95% by weight of a branched poly-α-olefin.

The first object of the present invention is to provide a high-performance traction fluid having a high traction coefficient. The second object of the present invention is to provide a traction fluid which is not only economical but also readily available and easily applicable to transmissions.

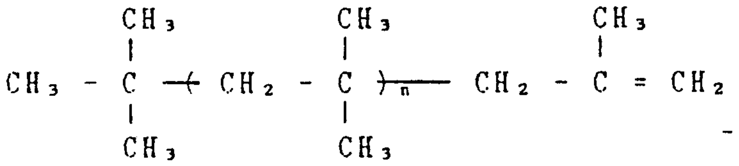
The traction fluid of the present invention comprises a base oil comprised of two components, i.e., component A comprised of a diester or its derivative, and a specific amount of a component B comprised of a branched poly-α-olefin.

In the present invention component A is a diester or its derivative having two cyclohexyl rings and is represented by the above-mentioned structural formula. A′ of the ester linkage is -COO- or -OOC-, and the number. n. of the carbon atoms in the hydrocarbon skeleton is 1 to 10, preferably 1 to 4. When n is zero, the traction coefficient is low while when n is 11 or more the viscosity is unfavorably high. This diester or derivative thereof can be prepared by the methods stated below and has a viscosity of 5 to 50 cst, preferably 7 to 30 cst at 40° C, and 1 to 10 cst, preferably 2 to 6 cst, at 100° C. The derivative includes an amino compound, an ether compound, etc.

The component A can be prepared by any of the following methods. The first method comprises an esterification reaction of a dihydric alcohol with a cyclohexanecarboxylic acid compound. The dihydric alcohol is selected from the group consisting of those having 1 to 10 carbon atoms, preferably 1 to 4 carbon atoms. Specifically, examples of the dihydric alcohol include ethylene glycol. 1,3-propanediol, 1,3-butanediol and 1,4-butanediol. Examples of the cyclohexanecarboxylic acid compound include, besides cyclohexanecarboxylic acid, those having an alkyl group with 1 to 8 carbon atoms, e.g., methylcyclohexanecarboxylic acid, ethylcyclohexanecarboxylic acid, etc. Cyclohexanecarboxylic acid is particularly preferred. The esterification reaction is conducted with an alcohol/acid molar ratio of 1:2, or in the presence of an excess amount of the acid. The former method requires the use of a catalyst and has the additional disadvantage that a monoalcohol is produced as the by-product. Therefore, it is preferred that the esterification reaction be conducted in the presence of an excess amount of the acid. Specifically, 1 mol of the dihydric alcohol is reacted with the acid in 2 to 5-fold mol excess (particularly preferred is a 2.5 to 4-fold mol excess). The reaction temperature is about 150 to 250° C, preferably 170 to 230° C, and the reaction time is 10 to 40 hr, preferably 15 to 25 hr. Although the esterification reaction may be conducted under either elevated or reduced pressures, it is preferred that the reaction is conducted at atmospheric pressure from the standpoint of ease of reaction operation. Under this condition the excess acid serves as a catalyst. An alkylbenzene such as xylene or toluene can be added in a suitable amount as a solvent. The addition of the solvent enables the reaction and temperature to be easily controlled. As the reaction proceeds the water which forms during the reaction evaporates. The reaction is terminated when the amount of the water reaches, on a mole basis, twice that of the alcohol. The excess acid is neutralized with an aqueous alkaline solution and removed by washing with water. When an acid which is difficult to extract with an alkali washing is used the reaction is conducted using the acid in an amount of 2 to 2.5-fold mol excess over the alcohol in the presence of a catalyst. Examples of the catalyst include phosphoric acid, p-toluenesulfonic acid, and sulfuric acid. The most preferable catalyst is phosphoric acid because it enhances the reaction rate and increases the yield of the ester. The reaction product is finally distilled under reduced pressure to remove water and the solvent thereby obtaining the diester compound of the present invention.

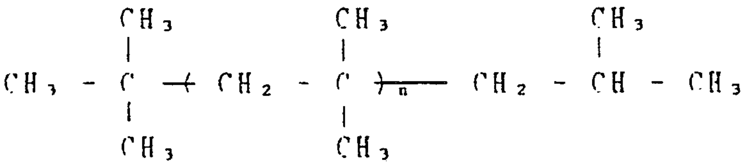
The second method of producing the component A of the present invention comprises esterification of a cyclohexanol compound with a dicarboxylic acid having 3 to 12 carbon atoms. Examples of the cyclohexanol compound include, besides cyclohexanol, those having an alkyl group with 1 to 8 carbon atoms, e.g., methylcyclohexanol and tertbutylcyclohexanol. Cyclohexanol is particularly preferred. The dicarboxylic acid includes one having 3 to 12 carbon atoms in its main chain, preferably one having 3 to 6 carbon atoms in its main chain. Examples of the dicarboxylic acid include malonic acid, succinic acid and glutaric acid. The esterification reaction is conducted in an alcohol/acid molar ratio of 2:1 or in the presence of an excess amount of the alcohol. In the former method, there is a possibility of forming a monocarboxylic acid as the by-product. Therefore, it is preferred that the esterification reaction is conducted in the presence of an excess amount of the alcohol. Specifically, 1 mol of the dicarboxylic acid is reacted with the alcohol in 2.5 to 5-fold mol excess. The reaction temperature is about 150 to 250° C, preferably 170 to 230° C, and the reaction time is 10 to 40 hr, preferably 15 to 25 hr. Although the esterification reaction may be conducted under either elevated or reduced pressures, it is preferred that the reaction be conducted at atmospheric pressure from the standpoint of ease of reaction operation. An alkylbenzene such as xylene or toluene can be added in a suitable amount as a solvent. The addition of the solvent enables the reaction temperature to be easily controlled. As the reaction proceeds the water which has been formed during the reaction evaporates. The reaction is terminated when the amount of the water reaches twice by mol that of the alcohol. Phosphoric acid, p-toluenesulfonic acid or sulfuric acid can be used as a catalyst. The most preferable catalyst is phosphoric acid because it enhances the reaction rate and increases the yield of the ester. The reaction product is finally distilled under reduced pressure to remove the water, solvent and excess alcohol thereby obtaining the diester compound of the present invention.

The poly-α-olefin component B has either a quaternary carbon atom or a tertiary carbon atom in its main chain and is a polymer of an α-olefin having 3 to 5 carbon atoms or the hydrogenation product thereof. Examples of the poly-α-olefin include polypropylene, polybutene, polyisobutylene and polypentene and the hydrogenation products thereof. Polybutene, polyisobutylene and hydrogenation products thereof are particularly preferred. The polyisobutylene is represented by the following structural formula:



$$CH_3-\underset{CH_3}{\overset{CH_3}{C}}\left(CH_2-\underset{CH_3}{\overset{CH_3}{C}}\right)_n CH_2-\overset{CH_3}{C}=CH_2$$

The hydrogenation product of the polyisobutylene is represented by the following structural formula:



$$CH_3-\underset{CH_3}{\overset{CH_3}{C}}\left(CH_2-\underset{CH_3}{\overset{CH_3}{C}}\right)_n CH_2-\overset{CH_3}{CH}-CH_3$$

In the above-mentioned formulae, the degree of polymerization, n, is 6 to 200.

Although the polybutene and polyisobutylene used may be commercially available ones, they may also be produced by conventional and well known polymerization methods. The hydrogenation product thereof is produced by reacting polyisobutylene or the like in the presence of hydrogen. The molecular weight of the poly-α-olefin is preferably in the range of 500 to 10,000, more preferably in the range of 900 to 5,000. The molecular weight can be adjusted by suitable methods such as decomposition of a poly-α-olefin having a high molecular weight and mixing of poly-α-olefins having different molecular weights. Although an α-olefin copolymer (OCP) is a type of a poly-α-olefin, it is unsuited for use as the component B in the present invention. This is because OCP is obtained by polymerization of two or more α-olefins and has such a structure that these α-olefins are irregularly linked, as opposed to the polybutene etc. of the present invention which have a regular gem-dialkyl structure.

The component A in the present invention, e.g., a diester of succinic acid with cyclohexanol, exhibits a traction coefficient of 0.102 to 0.106, while the component B, e.g., polybutene, exhibits a traction coefficient of 0.075 to 0.085.

Since the component A in the present invention exhibits a high traction coefficient, the use of the component A alone in a traction drive device results in a high performance. However, a further improved traction fluid can be obtained by blending the component A with 0.1 to 95% by weight, particularly 10 to 70% by weight, of the poly-α-olefin of the component B. Namely, although the component B has a lower traction coefficient than that of the component A, the gem-dialkyl group in the component B cooperates with the cyclohexyl ring in the component A to exhibit a synergistic effect (with respect to improvement in the traction coefficient). Further, since the component B is inexpensive and exhibits excellent viscosity characteristics, a traction fluid can be economically obtained by blending component A with 0.1 to 95% by weight of component B without lowering the traction coefficient.

Various additives may also be added to the traction fluid of the present invention depending upon their applications. Specifically, when the traction device undergoes a high temperature and a large load at least one additive selected from among an antioxidant, a wear inhibitor and a corrosion inhibitor may be added in an amount of 0.01 to 5% by weight. Similarly, when a high viscosity index is required a known viscosity index improver is suitably added in an amount of 1 to 10% by weight. However, since the use of polymethacrylates or olefin copolymers lowers the traction coefficient, it is preferred that, if present, they are used in an amount of 4% or less by weight.

The term "traction fluid" as used in the present invention is intended to mean a fluid for use in devices which transmit a rotational torque through point contact or line contact, or for use in transmissions having a similar structure. The traction fluid of the present invention exhibits a traction coefficient higher than those of conventionally known fluids, i.e., exhibits a traction coefficient 5 to 15% higher than those of the conventional fluids, although the value varies depending on the viscosity. Therefore, the traction fluid of the present invention can be advantageously used for relatively low power drive transmissions including internal combustion engines of small passenger cars, spinning machines and food producing machines, as well as large power drive transmissions such as industrial machines, etc.

The traction fluid of the present invention is remarkably superior in its traction coefficient relative to conventional fluids. The reason why the traction fluid of the present invention exhibits a high traction coefficient is not yet fully understood. However, basically, the reason is believed to reside in the unique molecular structure of the traction fluid of the present invention.

The component A of the traction fluid of the present invention comprises a diester having two cyclohexyl rings in its molecule. The two ester linkages bring about an interdipolar force between the molecules. It is believed that the interdipolar force serves to bring the fluid into a stable glassy state under high load conditions, thereby increasing the shear strength. Furthermore, the component B in the traction fluid of the present invention has a quaternary carbon atom of the gem-dialkyl type. Therefore when the traction device is under high load conditions the cyclohexyl rings are firmly engaged, like gears, with the gem-dialkyl portions of the quaternary carbon of the component B, while when the device is released from the load this engagement is quickly broken thereby causing fluidization.

MOST PREFERRED EMBODIMENTS OF THE PRESENT INVENTION

EXAMPLES 1-13

Diester A, of the present invention was synthesized by the following method.

First, 250 g of cyclohexanol and 104 g of malonic acid (i.e., 0.4 mol per mol of cyclohexanol) were charged into a reactor, and phosphoric acid was added in an amount of 1% by weight based on the total weight of the reactants. The reactor was heated at 180° C. The contents of the reactor were allowed to react at a temperature in the range of 180° C to 210° C under atomospheric pressure. The reaction was stopped at a point when the water generated during the reaction amounted to twice, by mol. of the amount of the malonic acid. The reaction mixture was washed with an alkaline solution to remove unreacted compounds from the mixture of the reaction products, i.e., a diester of cyclohexanol with malonic acid, the unreacted cyclohexanol and phosphoric acid, followed by vacuum distillation, thereby isolating a pure diester (A_1).

Using the same method as described above diesters A_2 and A_3 of the present invention were synthesized using the following raw materials:

A_2 ..... ethylene glycol and cyclohexanecarboxylic acid (in excess acid)

A_3 ..... succinic acid and cyclohexanol

The diesters thus produced were each blended with polybutene having an average molecular weight of 900 to 2350 followed by the measurement of traction coefficient. The conditions of the measurement of the traction coefficient were as follows:

measuring apparatus: Soda-type four-roller traction testing machine

test conditions: a fluid temperature of 20° C; a roller temperature of 30° C; a mean Hertzian pressure of 1.2

GPa; a rolling velocity of 3.6 m/s; and a slipping ratio of 3.0%.

The traction fluids of the present invention were found to be remarkably superior in traction performance to the conventional fluids as shown in Table 1.

COMPARATIVE EXAMPLES 1 TO 9

The traction coefficients of the following traction fluids were measured under the same conditions as those used in the above examples: a traction fluid consisting of 100% by weight of the component B; traction fluids obtained by blending the component A_1 to A_3 with 5 to 30% by weight of OCP or PMA; and a commercially available traction fluid (Santotrack ®).

The results are shown in Table 1. As can be seen from Table 1, all the comparative samples exhibited traction coefficients 5 to 15% smaller than those of the traction fluids of the present invention. Incidentally, OCP is an olefin copolymer. Specifically, an ethylene-propylene copolymer having an average molecular weight of 150,000 to 300,000 was used as OCP. PMA is a polymethyacrylate. Specifically, a polymer having an average molecular weight of 50,000 to 300,000 was used as PMA.

Table 1

	A		B		Viscosity (cst)		Viscosity index	Traction coefficient
	Loadings %		M.W.	Loadings %	40°C	100°C		
Reference 1	A_1		-	0	7.4	2.05	55	0.102
2	A_2		-	0	12.2	2.97	93	0.091
3	A_3		-	0	23.5	3.86	4.9	0.104
Example 1	A_1	90	900	40	60.9	8.69	116	0.112
2	"	80	1260	20	23.2	4.69	122	0.112
3	"	70	"	30	46.9	7.65	130	0.113
4	"	60	"	40	100.0	12.60	119	0.113
5	"	90	2350	10	16.1	4.8	252	0.111
6	A_2	70	1260	30	78.2	10.80	126	0.102
7	"	60	"	40	155.0	16.90	117	0.103
8	A_3	80	900	20	50.1	6.53	73	0.105
9	"	90	1260	10	39.7	5.75	78	0.106
10	"	80	"	20	71.9	8.60	90	0.108
11	"	70	"	30	123.0	12.89	91	0.112
12	"	60	"	40	269.3	20.13	86	0.109
13	"	80	2350	20	125.9	13.61	104	0.111
Comp. Ex. 1		0	900	100	11600	240	*108	0.081
2		0	1260	100	32000	630	*155	0.080
3	A_1	96	OCP	5	17.4	4.51	187	0.100
4	"	90	"	10	51.6	12.4	248	0.098
5	"	96	PMA	10	16.7	4.32	180	0.098
6	"	90	"	30	71.4	15.7	235	0.092
7	A_2	90	OCP	10	48.2	16.9	366	0.085
8	A'_3	90	"	10	95.3	23.5	277	0.099
9	Santotrack®				13.8	2.99	46	0.087
Note:								

* values obtained through calculation using an equation with respect to a kinetic viscosity of 17 to 43 cst.

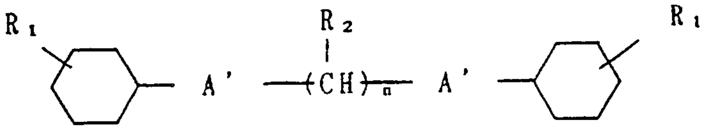
INDUSTRIAL APPLICABILITY

The present invention relates to a traction fluid which comprises a component A comprised of a diester or the like having two cyclohexyl rings and linear-chain hydrocarbons as the skeleton and a specific amount of a component B comprised of a branched poly-α-olefin, and which exhibits not only an extremely high traction coefficient, but also is inexpensive and exhibits excellent viscosity characteristics.

Therefore, the use of this traction fluid in a power transmission device, particularly a traction drive device, leads to a remarkable increase in shearing force under a high load, and at the same time enables reduction in size of the device and reduced cost of the device.

Claims

1. A traction fluid characterized by being a blend of a diester or its derivative represented by the general formula



wherein A′ is an ester linkage of -COO- or -OOC-, n is an integer of 1 to 10, each R_1 is independently selected from a hydrogen atom and alkyl groups having 1 to 8 carbon atoms, and R_2 is independently selected from a hydrogen atom and alkyl groups having 1 to 3 carbon atoms; with 0.1 to 95% by weight of a branched poly-α-olefin.

2. A traction fluid according to Claim 1 wherein said poly-α-olefin is polybutene.

3. A traction fluid according to Claim 1 which contains from 10 to 70% by weight of said poly-α-olefin.

4. A traction fluid of Claim 1 wherein each R_1 is the same or a different.group selected from a hydrogen atom and alkyl groups having 1 to 4 carbon atoms.

5. A traction fluid according to Claim 1 wherein said poly-α-olefin has an average molecular weight of 500 to 10,000.

6. A traction fluid of Claim 1 wherein n is an integer of 1 to 4.

7. A traction fluid of Claim 1 wherein R_2 is either hydrogen or a methyl group.

INTERNATIONAL SEARCH REPORT

International Application No PCT/JP87/00565

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [3]

According to International Patent Classification (IPC) or to both National Classification and IPC

Int.Cl^{4} C10M111/04, C10N40:04

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
IPC	C10M111/04, 105/38, 107/02-107/10 C10N40:04

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [5]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category *	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X	JP, A, 59-191797 (Nippon Petrochemicals Co., Ltd.) 30 October 1984 (30. 10. 84) Column 1, lines 5 to 11, column 8, line 4 to column 9, line 11, table 1 (Family: none)	1-7
Y	JP, A, 59-68397 (Maruzen Oil Co., Ltd.) 18 April 1984 (18. 04. 84) Column 1, lines 5 to 11, column 4, line 16 to column 6, line 11 (Family: none)	1-7
Y	JP, A, 61-19697 (Nippon Steel Chemical Co., Ltd.) 28 January 1986 (28. 01. 86) Column 1, line 5 to column 2, line 3, column 8, line 1 to column 9, line 4 (Family: none)	1-7
Y	JP, A, 54-96667 (Bayer A.G.) 31 July 1979 (31. 07. 79) Column 1, line 6 to column 2, line 1, column 4, line 12 to column 6, line 3,	1-7

* Special categories of cited documents: [15]

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

IV. CERTIFICATION

Date of the Actual Completion of the International Search [2] September 17, 1987 (17. 09. 87)	Date of Mailing of this International Search Report [2] September 28, 1987 (28. 09. 87)
International Searching Authority [1] Japanese Patent Office	Signature of Authorized Officer [20]

International Application No. PCT/JP87/00565

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET		
	column 18, line 1 to column 19, line 14 & DE, A1, 2758780 & EP, A1, 3032 & US, A, 4212816 & EP, B1, 3032	
Y	JP, A, 53-127970 (Nippon Oil and Fats Co., Ltd.) 8 November 1978 (08. 11. 78) Column 1, lines 5 to 17, column 5, line 8 to column 6, line 20 (Family: none)	1-7
Y	JP, A, 47-2229 (Sun Oil Company) 3 February 1972 (03. 02. 72)	1-7

V.☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

This international search report has not been established in respect of certain claims under Article 17(2) (a) for the following reasons:

1.☐ Claim numbers............ because they relate to subject matter [12] not required to be searched by this Authority, namely:

2.☐ Claim numbers............ because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out [13], specifically:

VI.☐ OBSERVATIONS WHERE UNITY OF INVENTION IS LACKING[11]

This International Searching Authority found multiple inventions in this international application as follows:

1.☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims of the international application.

2.☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims of the international application for which fees were paid, specifically claims:

3.☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claim numbers:

4.☐ As all searchable claims could be searched without effort justifying an additional fee, the International Searching Authority did not invite payment of any additional fee.

Remark on Protest

☐ The additional search fees were accompanied by applicant's protest.

☐ No protest accompanied the payment of additional search fees.

International Application No. PCT/JP87/00565

FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET

	Column 9, line 14 to column 12, line 10 (Family: none)	
Y	JP, A, 61-9497 (Nippon Oil Co., Ltd.) 17 January 1986 (17. 01. 86) Column 1, lines 5 to 16, column 7, lines 5 to 19 (Family: none)	1-7

V.☐ OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE[10]

This international search report has not been established in respect of certain claims under Article 17(2) (a) for the following reasons:

1.☐ Claim numbers............ because they relate to subject matter [12] not required to be searched by this Authority, namely:

2.☐ Claim numbers............ because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out [13], specifically:

VI.☐ OBSERVATIONS WHERE UNITY OF INVENTION IS LACKING[11]

This International Searching Authority found multiple inventions in this international application as follows:

1.☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims of the international application.

2.☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims of the international application for which fees were paid, specifically claims:

3.☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claim numbers:

4.☐ As all searchable claims could be searched without effort justifying an additional fee, the International Searching Authority did not invite payment of any additional fee.

Remark on Protest

☐ The additional search fees were accompanied by applicant's protest.

☐ No protest accompanied the payment of additional search fees.