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(54) **IMPROVED TRACTION FUID.**

(57) An improved traction fluid characterized by being a blend of (i) an ester or its derivative, in which the ester is represented by the general formula:

wherein A' is an ester linkage of -COO- or -OOC-, n is an integer of 1 to 14, R_1 is a member selected from among the hydrogen atom and alkyl groups having 1 to 8 carbon atoms, and R_2s are the same or different and are a member selected from among a hydrogen atom and alkyl groups having from 1 to 3 carbon atoms, with (ii) from 0.1 to 95 % by weight of a hydrocarbon polymer or polymeric ester.

This fluid is advantageously used for power drive transmissions, particularly for traction drive devices.

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IMPROVED TRACTION FLUID

FIELD OF THE INVENTION

This invention relates to an improved traction fluid. More particularly, it relates to a traction fluid comprising a blend of an ester compound or its derivative in which a cyclohexyl ring is connected to a linear-chain hydrocarbon through an ester linkage, and a hydrocarbon polymer or polymeric ester as the base oil.

BACKGROUND OF THE INVENTION

Traction drive power transmissions which transmit power to a driven part through a traction drive mechanism in automobiles and industrial machinery have recently attracted attention, and in recent years research and development in this area has been progressing. A traction drive mechanism is a power transmitting mechanism using a rolling friction. Unlike conventional drive mechanisms, it does not use any gears. This enables reduced vibrations and noise, as well as providing smooth speed changes in highspeed rotation. An important goal in the automobile industry is to improve automobile fuel economy. It has been suggested that if the traction drive were to be used for automobile transmissions, in order to convert the transmission to the continuous variable-speed transmission, that fuel consumption could be reduced by at least 20% over conventional transmission systems. This is because the drive can always be in the optimum fuel consumption region of an engine. Recent studies have resulted in developing materials having high fatigue resistance, and in a theoretical analysis of traction mechanisms. As regards traction fluids, 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 that 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.

A traction fluid must be comprised of a lubricating oil having a high traction coefficient. It is known that a traction fluid possessing a molecular structure having a naphthene ring exhibits such a high performance. "Santotrack®," manufactured by the Monsanto Chemical Company, is widely known as a commercially available traction fluid. Japanese Patent Publication No. 47-35763 discloses di(cyclohexyl)alkane and dicyclohexane as traction fluids having a naphthene ring. This patent publication discloses that a fluid obtained by incorporating the above-mentioned alkane compound in perhydrogenated (α-methyl)styrene polymer, hydrindane compound or the like, has a high traction coefficient. Further, Japanese Patent Laid Open No. 59-191797 discloses a traction fluid containing an ester compound having a naphthene ring. It discloses that an ester obtained by the hydrogenation of the aromatic nucleus of dicyclohexyl cyclohexanedicarboxylate or dicyclohexyl phthalate is preferred as the traction fluid.

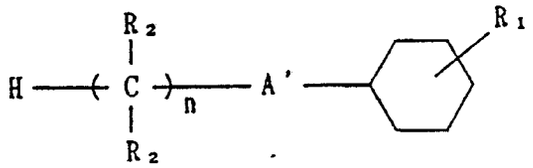
As mentioned above, in recent years there has been progress in the development of continuous variable-speed transmissions in the automobile industry. The higher the traction coefficient of the traction fluid, the larger the allowable transmission force in the same device. This helps reduce the size of the entire device, with a corresponding reduction in polluting exhaust gas emissions. Therefore, there is a need for a fluid having a traction coefficient which is as high as possible. However, the use of the currently commercially available traction fluid exhibiting the highest performance of all such currently commercially available fluids in such a traction drive device performs unsatisfactorily in respect to the traction coefficient. Further, such a traction fluid is rather costly. The traction fluid which has been proposed in Japanese Patent Publication No. 46-35763 contains α-methylstyrene polymer or its analogue as a component and, therefore, is also has an unsatisfactory performance and cost.

DISCLOSURE OF THE INVENTION

The inventors have made extensive and intensive studies to develop a traction fluid which not only exhibits a high traction coefficient, but is also economical. As a result the inventors found that the

combination of a specific amount of a hydrocarbon polymer, or polymeric ester and an ester compound or its derivative in which a cyclohexyl ring is connected to a linear-chain hydrocarbon through an ester linkage, can provide a high-performance and economical base oil fluid. This invention is based on this finding.

This invention relates to a traction fluid characterized by being a blend of (i) an ester or its derivative, in which the ester is represented by the general formula:



wherein A′ is an ester linkage of -COO- or -OOC-, n is an integer of 1 to 14, R_1 is a member selected from among a hydrogen atom and alkyl groups having 1 to 8 carbon atoms, and R_2s are the same or different and are a member selected from among a hydrogen atom and alkyl groups having 1 to 3 carbon atoms, with (ii) from 0.1 to 95 % by weight of a hydrocarbon polymer or polymeric ester

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

The traction fluid of this invention comprises a base oil comprised of two components, i.e., component A comprised of an ester or its derivative, and a specific amount of component B comprised of a hydrocarbon polymer or polymeric ester

The component A of this invention is an ester compound or its derivative in which a cyclohexyl ring is connected to a linear-chain hydrocarbon through an ester linkage and has the above-mentioned structural formula. A′ of the ester linkage is -COO- or -OCC-, and the number, n, of the carbon atoms in the hydrocarbon skeleton, is 1 to 14, preferably 2 to 10. When n is zero, the traction coefficient is low, while when n is 15 or more the viscosity is unfavorably high. This ester or derivative thereof prepared by the methods stated below has a viscosity of 3 to 150 cst, preferably 4 to 120 cst, at 40 °C, and 1 to 20 cst, preferably 1 to 16 cst, at 100 °C. Examples of the derivative of the ester include an amino compound, a halide compound, and an ether compound.

The component A can be prepared by any of the following methods: The first method comprises an esterification reaction of a monohydric alcohol with a cyclohexanecarboxylic acid compound. The monohydric alcohol has 1 to 14 carbon atoms, preferably 2 to 10 carbon atoms, in its main chain. Specifically, examples of the monohydric alcohol include ethanol, propanol, 2-propanol, butanol, 2-methyl-2-propanol, 3,3,5-trimethyl-1-hexanol, and 3,3,5,5-tetramethyl-1-hexanol. 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 in the presence of an excess amount of the alcohol or acid. When the esterification is conducted under excess acid conditions, 1 mol of the monohydric alcohol compound is reacted with 1 to 2 mol (particularly preferably 1.2 to 1.8 mol) of the acid. 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 preferable to do so at atmospheric pressure, from the standpoint of ease of reaction operation. Under these conditions 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 temperature to be easily controlled. As the reaction proceeds, water formed during the reaction evaporates. The reaction is terminated when an equimolar amount of the water, with respect to the alcohol, has evaporated. 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 equimolar amounts of the acid and 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. Finally, the reaction product is distilled under reduced pressure to remove water and the solvent, thereby obtaining the ester compound of this invention.

When the esterification reaction is conducted under excess alcohol conditions, 1 mol of the cyclohexanecarboxylic acid compound is reacted with1 to 2 mol (particularly preferably 1.2 to 1.8 mol) of the alcohol. 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 preferable that it be done at atmospheric pressure, from the standpoint of ease of reaction operation. Under these conditions 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 temperature to be easily controlled. As the reaction proceeds, water which has been formed during the reaction evaporates. The reaction is terminated when an equimolar amount of the water, with respect to the acid, has evaporated. Examples of the catalysts include phosphoric acid, p-toluenesulfonic acid and sulfuric acid. The most preferred catalyst is phosphoric acid, because it enhances the reaction rate and increases the yield of the ester. After the reaction is terminated the reaction product is neutralized with an aqueous alkaline solution and is washed with water to remove the unreacted acid and catalyst. Finally, the reaction product is distilled under reduced pressure to remove water, the solvent, and the excess alcohol, thereby obtaining the ester compound of this invention.

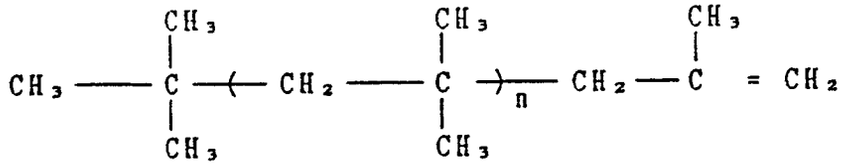
The second method of producing the component A of this invention comprises esterification of a cyclohexanol compound with a carboxylic acid having 2 to 15 carbon atoms in its main chain. Examples of the cyclohexanol compounds include, besides cyclohexanol, those having an alkyl group with 1 to 8 carbon atoms, e.g., methylcyclohexanol, ethylcyclohexanol, and tert-butylcyclohexanol. Cyclohexanol is particularly preferred. The dicarboxylic acid includes one having 2 to 15 carbon atoms in its main chain, preferably one having 3 to 11 carbon atoms. Examples of the carboxylic acids include acetic, propionic, butyric, lauric, trimethylhexanoic, and tetramethylhexanoic acids. The esterification reaction is conducted in the presence of an excess amount of the acid or alkali. When the esterification reaction is conducted under excess acid conditions, 1 mol of a cyclohexanol compound is reacted with 1 to 2 mol (particularly preferably 1.2 to 1.8 mol) of the acid. 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 this esterification reaction may be conducted under either elevated or reduced pressures, it is preferable to do so at atmospheric pressure, from the standpoint of ease of reaction operation. Phosphoric acid, p-toluenesulfonic acid, or sulfuric acid, is used as the catalyst. The most preferable catalyst is phosphoric acid, because it enhances the reaction rate and increases the yield of the ester. 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, water which has been formed during the reaction evaporates. The reaction is terminated when an equimolar amount of the water, with respect to the alcohol, has evaporated. The catalyst and the excess acid are removed by neutralizing them with an aqueous alkaline solution and washing them with water. The reaction product is finally distilled, under reduced pressure, to remove the water and solvent, thereby obtaining the ester compound of this invention.

When the esterification reaction is conducted under excess alcohol conditions, 1 mol of the carboxylic acid is reacted with 1 to 2 mol (particularly preferably 1.2 to 1.8 mol) of the cyclohexanol compound. 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 it 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, water which has been formed during the reaction evaporates. The reaction is terminated when an equimolar amount of the water, with respect to the carboxylic acid, has evaporated. Examples of the catalyst include phosphoric acid, p-toluenesulfonic acid, and sulfuric acid. The most preferred catalyst is phosphoric acid, because it enhances the reaction rate and increases the yield of the ester. After termination of the reaction, the reaction product is neutralized with an aqueous alkaline solution, and it is then washed with water to remove the catalyst. Finally, the reaction product is distilled under reduced pressure to remove the water, the solvent, and the excess alcohol, thereby obtaining the ester compound of this invention.

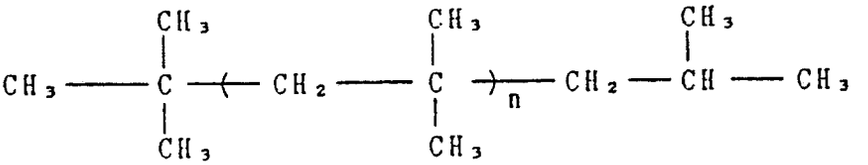
Regarding component B, the hydrocarbon, polymer is a polyolefin, a hydrocarbon polymeric compound having a naphthene ring, or an alkylstyrene polymer, while the polymeric ester is a polymer of an ester compound having a side chain of an alkyl group and/or cyclohexyl ring. Examples of the polyolefin include a poly-α-olefin, an olefin copolymer, a polymer obtained by saturating the unsaturated bonds of one of the above polymers with hydrogen, and a modified polymer obtained by modifying one of the above polymers with a small amount of a modifier. Examples of the modifier include alkylcarboxylic acids, alcohols, and amines. Among the above-mentioned polymers, a poly-α-olefin is particularly preferred.

The poly-α-olefin, which is a preferred component B, has a 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. Particularly preferred are polybutene and polyisobutylene and the hydrogenation products thereof. The polyisobutylene is represented by the following structural formula:



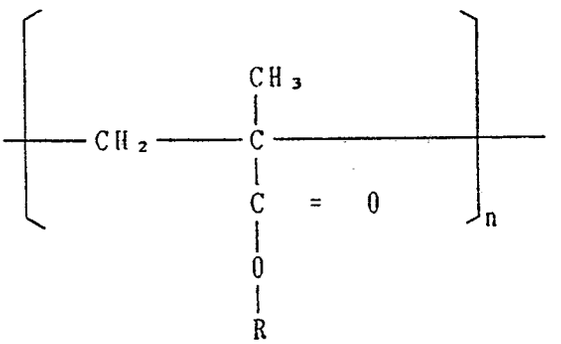
The hydrogenation product of the polyisobutylene is represented by the following structural formula:



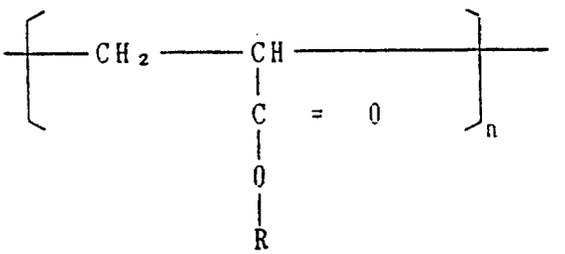
In the above-mentioned formula, the degree of polymerization, n, is 6 to 200.

Although the polybutene and polyisobutylene may be commercially available ones, they may also be produced by conventional 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 with poly-α-olefins having different molecular weights. The olefin copolymer (OCP) can be obtained by polymerization of two or more olefins selected from among ethylene, propylene, butene, pentene, and styrene. OCP has such a structure that the olefins are irregularly linked with each other, as opposed to a poly-α-olefin, such as polybutene, which has a regular gem-dialkyl structure.

Examples of the polymeric ester useful as component B include polymethacrylate or its derivative represented by the following general formula:



wherein R is a hydrocarbon group having 1 to 18 carbon atoms and n is an integer of 100 to 5,000, and polyacrylate or its derivative represented by the general formula:



wherein R is a hydrocarbon group having 1 to 18 carbon atoms and n is an integer of 100 to 5,000. A preferred esteric polymer is polycyclohexyl acrylate or polycyclohexyl methacrylate having the above general formula in which R is a cyclohexyl ring having 6 to 12 carbon atoms and n is an integer of 200 to 250.

Component A in this invention, e.g., an ester of 3,5,5-trimethylhexanoic acid with cyclohexanol, exhibits a traction coefficient of 0.080 to 0.090, while component B, e.g., polybutene, exhibits a traction coefficient of 0.075 to 0.085.

Both components A and B of this invention have a lower traction coefficient than those of commercially available traction fluids. Therefore, they cannot exhibit a high performance if they are used singly or individually in a traction drive device. However, an excellent traction fluid can be obtained by blending component A with 0.1 to 95% by weight, particularly preferably 10 to 70% by weight, of component B, which is comprised of a hydrocarbon polymer or polymeric ester Specifically, a hydrocarbon group such as an alkyl group of component B cooperates with the cyclohexyl ring in component A to exhibit a synergistic effect (in respect to improvement of the traction coefficient). Further, since component B is inexpensive and exhibits excellent viscosity characteristics, a traction fluid exhibiting an enhanced traction coefficient can be economically obtained by blending the component A with 0.1 to 95% by weight of the component B.

Various additives may also be added to the traction fluid of this invention, depending on its applications. Specifically, when the traction device is subjected to a high temperatures 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.

The term "traction fluid" as used in this invention is intended to mean a fluid for use in devices which transmit a rotational torque through spot contact or line contact, or for use in transmissions having a similar structure. The traction fluid of this invention exhibits a traction coefficient higher than those of conventionally known fluids, i.e., exhibits a traction coefficient 1 to 10 % higher than those of the conventional fluids, although the value varies depending on the viscosity. Therefore, the traction fluid of this invention can be advantageously used for relatively low power drive transmissions, including internal combustion engines of small passenger cars, spinning machines, and food processing machines, as well as for large power drive transmissions such as industrial machines, etc.

The traction fluid of this invention exhibits a remarkably superior traction coefficient compared to conventional fluids. The reason why it does so is not yet fully understood. Basically, however, the reason is believed to reside in the unique molecular structure of the traction fluid of this invention.

The component A of the traction fluid of this invention comprises an ester having a cyclohexyl ring in its molecule. The ester linkage brings about an interdipolar force between the molecules. It is believed that this interdipolar force brings the fluid into a stable glassy state under high load conditions, thereby increasing the shearing force. Further, component B of the traction fluid of this invention has a hydrocarbon group such as an alkyl group. Therefore, when the traction device is under high load conditions the cyclohexyl ring in the component A is firmly engaged, like gears, with the alkyl group in component B, while when the device is released from the load this engagement is quickly broken, thereby causing fluidization.

THE OPTIMUM EMBODIMENTS OF THE INVENTION

EXAMPLES 1-18

Ester A_1 of this invention was synthesized by the following method: First, 150.2 g of cyclohexanol and 158.2 g of 3,5,5-trimethylhexanoic acid (wherein the mole ratio of the alcohol to the acid was 1.5:1) 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. Then the reactor was heated to 180 °C and the contents of the reactor were allowed to react at a temperature in the range of 170 °C to 200 °C under atmospheric pressure. The heating was stopped at a point when the water generated during the reaction amounted to the same number of moles as the above acid.

The reaction mixture was washed with an alkaline solution to remove unreacted compounds, i.e., cyclohexanol, and the phosphoric acid catalyst, from a mixture of a reaction product, i.e., an ester of cyclohexanol with trimethylhexanoic acid, the unreacted compounds, and phosphoric acid, followed by vacuum distillation, thereby isolating a pure diester A_1.

In the same manner as described above, the other esters of this invention, A_2 and A_3, were synthesized using the following materials:

A_2 ... 3,5,5-trimethyl-1-hexanol and cyclohexane carboxylic acid (no catalyst was used due to the surplus of acid.)

A_3 ... Lauric acid and cyclohexanol (Phosphoric acid catalyst was used.)

Next, the ester thus produced was blended with polybutene B_1 having an average molecular weight of 900 to 2350, an olefin copolymer (OCP) B_2* or an esteric polymer B_3**, followed by measurement of the traction coefficient. The measurement conditions of the traction coefficient were as follows:

* A copolymer of ethylene with propylene having an average molecular weight of 32.3×10^4

** A polycyclohexylacrylate having an average molecular weight of 5×10^4

Measurement equipment: Soda-type four roller traction test machine.

Testing 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 percent slipping ratio of 3.0%.

The traction fluid obtained by this invention was found to be remarkably superior in its traction performance to the conventional fluids, as shown in Table 1.

COMPARATIVE EXAMPLES 1 - 5

Traction fluids prepared from A_1 to A_3 components alone and B_1 component alone, and a commercially available traction fluid (Santotrack®), were used as the comparative examples. The traction coefficients of these comparative examples were measured under the same conditions as described above.

The results are shown in Table 1. As can be seen from it, all the comparative examples exhibited traction coefficients 1 to 10% smaller than that of the traction fluid of this invention.

Table 1

		A		B			Kinetic Viscosity(cst)		Viscosity index	Traction coefficient
		Loadings wt%		Average M.W.		Loadings wt%	40°C	100°C		
Example	1	A_1	75	B_1	900	25	16.1	4.04	157	0.095
	2	"	66	"	"	34	25.4	5.73	178	0.096
	3	"	82	"	1260	18	13.0	3.82	209	0.091
	4	"	74	"	"	26	23.8	5.39	173	0.095
	5	"	66	"	"	34	39.7	8.19	187	0.100
	6	"	58	"	"	42	101.0	12.98	125	0.096
	7	"	88	"	2350	12	12.8	4.05	249	0.093
	8	"	70	"	"	30	91.9	14.38	162	0.100
	9	A_2	57	B_1	900	43	45.9	8.59	168	0.090
	10	"	70	"	1260	30	39.1	7.18	149	0.089
	11	"	62	"	"	38	75.1	10.84	132	0.089
	12	"	74	"	2350	26	50.3	11.30	226	0.090
	13	A_3	66	B_1	1260	34	64.2	9.31	123	0.091
	14	A_1	95	B_2	323000	5	10.0	3.13	200	0.087
	15	A_2	95	"	"	5	9.8	3.27	238	0.078
	16	A_3	95	"	"	5	16.2	4.45	206	0.083
	17	A_1	95	B_3	50000	5	11.4	3.52	215	0.092
	18	A_2	90	"	"	10	12.3	4.01	264	0.089
Comp. Ex.	1	A_1	100	-		0	5.0	1.66	85	0.084
	2	A_2	"	-		0	5.1	1.77	121	0.076
	3	A_3	"	-		0	8.5	2.31	78	0.080
	4	-		B_1	900	100	11600	240	108	0.081
	5	Santotrack®		-		0	13.8	2.99	46	0.087

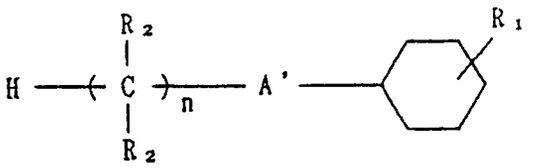
INDUSTRIAL AVAILABILITY

The traction fluid of this invention comprises a blend of a compound (component A) in which a cyclohexyl ring is connected to a linear-chain hydrocarbon through an ester linkage, and a specific amount of a hydrocarbon polymer or polymeric ester Such a traction fluid not only exhibits an extremely high traction coefficient but is also inexpensive and has excellent viscosity characteristics.

Therefore, the use of the traction fluid of this invention in a power transmission, particularly a traction drive device, results in a remarkable increase in shearing force under a high load. This enables the size of the device to be reduced, as well as reducing its costs.

Claims

1. An improved traction fluid characterized by being a blend of (i) an ester or its derivative, in which the ester is represented by the general formula:



wherein A' is an ester linkage of -COO- or -OOC-, n is an integer of 1 to 14, R_1 is a member selected from among a hydrogen atom and alkyl groups having 1 to 8 carbon atoms, and R_2s are the same or different and are a member selected from among a hydrogen atom and alkyl groups having from 1 to 3 carbon atoms, with (ii) from 0.1 to 95% by weight of a hydrocarbonic polymer or esteric polymer.

2. A fluid of claim 1 wherein the hydrocarbonic polymer is a polyolefin.

3. A fluid of claim 1 in which the hydrocarbonic polymer or esteric polymer is blended in an amount of 10 to 70 % by weight.

4. A fluid of claim 1 or 2 wherein the hydrocarbonic polymer is polybutene.

5. A fluid of claim 1 wherein R_1 of the ester is selected from hydrogen and alkyl groups having from 1 to 4 carbon atoms.

6. A fluid of claim 1 to 4 wherein the hydrocarbonic polymer or esteric polymer has an average molecular weight of from 500 to 10,000.

7. A fluid of claims 1-5 wherein n of the ester is an integer of 1 to 10.

8. A fluid of claim 1, 5, or 7 wherein R_2 of the ester is a hydrogen atom or a methyl group.

INTERNATIONAL SEARCH REPORT

International Application No PCT/JP87/00706

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] C10M105/34, 107/02, 107/28, 111/04, C10N40:04

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
IPC	C10M105/32, 105/34, 107/02-107/10, 107/28, 111/04, 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]
A	JP, A, 59-191797 (Nippon Petrochemicals Co., Ltd.) 30 October 1984 (30. 10. 84) Column 1, lines 5 to 15 (Family: none)	1-8
A	JP, A, 61-188495 (Toa Nenryo Kogyo Kabushiki Kaisha) 22 August 1986 (22. 08. 86) Column 1, line 5 to column 2, line 5 (Family: none)	1-8

* Special categories of cited documents: [16]

"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

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IV. CERTIFICATION

Date of the Actual Completion of the International Search [2]	Date of Mailing of this International Search Report [2]
December 10, 1987 (10.12.87)	December 21, 1987 (21.12.87)
International Searching Authority [1]	**Signature of Authorized Officer [20]**
Japanese Patent Office	