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(54) **Compound useful as detergent additive, compositions comprising a lubricating oil or hydrocarbon fuel, and method for maintaining the cleanliness of an internal combustion engine.**

(57) Aralkyl acid and polyamine are reacted to produce amide that is a detergent lubricant and fuel additive that is particularly effective to keep two-cycle engines clean.

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Compound Useful as Detergent Additive, Compositions Comprising a Lubricating Oil or Hydrocarbon Fuel, and Method for Maintaining the Cleanliness of an Internal Combustion Engine

This invention relates to additives for hydrocarbons suitable for use in an internal combustion engine. In one of its aspects, this invention relates to detergent additives for lubricants and hydrocarbon fuels.

It is common practice to add detergent compounds, detergent compositions, or detergent packages, i.e., a mixture of detergent compositions along with other additives, to lubricant and fuel compositions to prevent the deposition of solid materials on engine surfaces which come into contact with the lubricant or fuel compositions. The deposits formed in the engine interfere with the proper circulation of the lubricant and fuel compositions and also act as abrasives which can aggravate the wearing of engine parts.

It has also been found that some detergent additives that have been employed in lubricant and fuel compositions actually contribute to the formation of deposits in modern engines. It is, therefore, always of interest when compounds or compositions that provide improved detergent additive properties are discovered.

It is, therefore, an object of this invention to provide a detergent additive for internal combustion engine fuels and lubricants. It is another object of this invention to provide a method for retarding the depositing of solid materials on the surfaces of internal combustion engines. It is still another object of this invention to provide detergent fuel compositions and detergent lubricating compositions for internal combustion engines.

Other aspects, objects and the various advantages of this invention will become apparent upon reading the specification and the appended claims.

Statement of the Invention

In accordance with this invention, detergent additives for lubricants and fuels for use in internal combustion engines is provided. The detergent additive is those amides made by reaction between aralkyl acids and polyamines. The detergent lubricant and fuel additives are particularly effective to keep two-cycle, outboard engines clean.

In one of its embodiments, this invention provides a fuel or lubricant composition for use in internal combustion engines. The composition is a mixture of a fuel or lubricant with a detergent additive amide, produced as described above.

In still another embodiment of the invention a method is provided for maintaining the cleanliness of an internal combustion engine by the addition to the hydrocarbon fuel or lubricant for the engine of a detergent additive amide produced as described above.

Aralkyl acids that are suitable for use to synthesize the additive are those acids that can be prepared by alkylation of an aromatic hydrocarbon such as benzene, toluene, xylenes, or the like with an unsaturated fatty acid such as the mono-unsaturated acids lauroleic, myristoleic, palmitoleic, oleic, gadoleic, and the like, or with poly-unsaturated fatty acids such as linoleic, linolenic, or moroctic acids--C_{18} acids having two, three, and four double bonds, respectively. The aralkyl radical R of the acids (RCOOH) can contain from about 11-41 carbon atoms; preferably it will contain 17-29 carbon atoms. Examples of suitable aralkyl acids are diphenyllauric acid, triphenylpalmitic acid, phenyl stearic acid, and the like in which at least one aromatic radical is combined in the original fatty acid.

Polyamines that are suitable for use can be represented by the formula $NH_2(CH_2CH_2NH)_xH$ in which x is an integer between two and six. Examples are diethylenetriamine, triethylenetetramine, and tetraethylenepentamine. Especially preferred is the latter amine in which x is four.

The ratio in which aralkyl acid and polyamine are combined for reaction is about 0.2-1.0 moles of acid per equivalent of nitrogen. Preferably the ratio is between about 0.4-0.8 moles of acid per equivalent of nitrogen.

Production of amides by reaction between aralkyl acids and polyamines is accompanied by the liberation of water, and is facilitated by the use of temperatures above the normal boiling point of water. Reaction can be effected without using a solvent by operating under conditions in which water vapor is removed from the reactants with the aid of a stream of gas, preferably inert, such as nitrogen or argon. Alternatively the reaction can be effected in a solvent such as a hydrocarbon that permits operation under reflux at a temperature of about 100-200°C. Since one mole of water is produced per mole of amide, measurement of evolved water provides a convenient method to follow the extent of the reaction.

In addition to being effective to maintain the cleanliness of an outboard engine during extended operation, the additive of this invention is useful to prevent or reduce the formation of harmful carburetor and fuel intake system deposits when admixed with the fuel of any internal combustion engines. For this application the additive is used at a concentration of about 1-250 pounds per thousand barrels of fuel.

The additive is also useful with lubricant stocks, particularly solvent refined, paraffinic stock having a viscosity index of 100 or above and a viscosity at 210°F of about 39 to about 100 Saybolt Universal seconds (SUS), preferably about 45 to about 75 SUS. Other additives commonly used to formulate lubricants, such as viscosity index improvers, antioxidants, and the like can be used in formulation with the additive of this invention without destroying the effectiveness of the additives.

The following example illustrates the preparation of an amide that is subsequently shown to possess good detergent properties when incorporated into the lubricant for a two-cycle outboard engine.

Example

Amide was prepared by combining 2238 g (6.0 moles) of phenylstearic acid (Neofat L-PS from Armak Co.) and 378 g (2.0 moles) of tetraethylenepentamine in a flask fitted with a stirrer, a water-cooled reflux condenser, a Barrett water trap, and a heating mantle. After addition of 835 ml of xylene the mixture was heated to boiling and maintained at reflux for 3.2 hours during which interval 125 ml (6.9 moles) of water was collected. Water in excess of the expected 6.0 moles was presumably present as an impurity in the reactants. Solvent was removed from the product by distillation at 132°C kettle temperature at 7 torr pressure.

The solvent-free additive was incorporated into a lubricating oil blend at a concentration of 10.1 volume percent. The resulting mixture was combined with gasoline in the volume ratio 1:50, respectively, and tested for 100 hours in a 25 horsepower Johnson two-cycle outboard engine at wide open throttle (about 4800-4900 rpm) except for five minutes of idling at hourly intervals. The additive was evaluated by the Piston Varnish Test, a rating system developed by the Coordinating Research Council (CRC) that indicates the relative amount of varnish deposit on a piston. In this system a rating of 10 designates a clean piston and zero designates a very dirty one. This system is used by the Boating Industry Association to certify two-cycle lubricants for service TC-W. A satisfactory additive should produce a Piston Varnish rating of at least 9.0. The additive made and evaluated as described in this example received a 9.7 rating.

CLAIMS

1. A compound useful as a detergent additive for lubricants and fuels prepared by reacting aralkyl acid and polyamine to produce an amide.

2. A compound of claim 1 wherein said aralkyl acid has the general formula RCOOH in which R contains from about 11 to about 41 carbon atoms including at least 1 aromatic radical and said polyamine is represented by the formula $NH_2(CH_2CH_2NH)_xH$ in which x is an integer between 2 and 6.

3. A compound of claim 2 in which x is 4.

4. A compound of claim 3 in which the aralkyl acid is phenylstearic acid and the polyamine is tetraethylenepentamine.

5. A composition comprising a lubricating oil and the compound of one of claims 1 to 4, said compound being present in an amount effective as a lubricating oil additive.

6. A composition comprising a hydrocarbon suitable as fuel in an internal combustion engine and a compound of one of claims 1 to 4, said compound being present in an amount effective as a fuel detergent additive.

7. A method for maintaining the cleanliness of an internal combustion engine comprising the addition of a detergent additive of claim 1 to the hydrocarbon fuel or lubricant for the engine, said detergent being added in an amount effective to reduce engine deposits and using said hydrocarbon fuel or lubricant with detergent additive as fuel or lubricant in an internal combustion engine.

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EUROPEAN SEARCH REPORT

Application number
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DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim
X	US - A - 3 429 674 (D.I. HOKE) * Complete document * ----	1-7

CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)

C 10 L 1/22
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TECHNICAL FIELDS SEARCHED (Int.Cl. 3)

C 10 L 1/22
1/10
1/14
C 07 C 103/87
103/82

CATEGORY OF CITED DOCUMENTS

X: particularly relevant
A: technological background
O: non-written disclosure
P: intermediate document
T: theory or principle underlying the invention
E: conflicting application
D: document cited in the application
L: citation for other reasons

&: member of the same patent family, corresponding document

X The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
The Hague	02-07-1980	ROTSAERT

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