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(54) **Additive to improve combustion in internal combustion engines and boilers**

Additiv zur Verbesserung der Verbrennung in Verbrennungsmotoren und Heizkesseln

Additif pour améliorer la combustion dans des moteurs à combustion interne et des chaudières

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## Description

### Field of the invention

**[0001]** The present invention relates to novel compositions of ingredients that are used as additives for liquid fuels, to produce clear and stable solutions with the fuels that meet or exceed the specifications of the fuels and that substantially improve the combustion of the fuels, reducing consumption of the fuel and emissions of carbon monoxide (CO) and unburned hydrocarbons (HC), formation of fumes, particles and carbonaceous deposits. Specifically, the composition of the additives includes an aqueous solution of one or more amino-acids and alcohols having 1 to 5 carbon atoms, and optionally reaction products of acids and basic nitrogenated products, methyl esters of fatty acids and/or solvents. These compositions can also include additives for improving the octane and cetane numbers. As used in this specification the term "liquid fuel" means not only any hydrocarbon mixture for internal combustion engines but any burnable liquid matter such as crude oil, distillate from slate and the like.

**[0002]** The deterioration of the atmosphere due to emissions from automobiles and boilers has resulted in a massive effort to improve the quality of fuels. The basic problem is that internal combustion engines and boilers are inherently inefficient. Only a fraction of the fuel is actually used, while the rest is dissipated as heat or vibration or used as friction between the moving parts. Part of the fuel is not completely combusted and passes to the exhaust fumes as hydrocarbons or carbon monoxide, the two main atmospheric pollutants. In view of the millions of automobiles and boilers in operation worldwide, it seems obvious that small improvements in the efficiency of internal combustion engines and boilers can result in significant savings of fuel and considerable reduction of air pollution.

### Prior art

**[0003]** Much time and effort has been devoted to producing fuels for internal combustion engines and boilers providing significantly reduced emissions of toxic gases and volatile organic compounds without affecting the efficiency and performance of the engines and boilers.

**[0004]** It is well known among experts in the field that introducing oxygenated compounds in fossil fuels helps to improve the combustion and to reduce the emission of toxic compounds. Several efforts have been made to increase the amount of oxygen in the combustion chamber. For example, turbo chargers and auxiliary air injectors have often been used to increase the air supply to the engines. For example, addition of pure oxygen to the air/fuel mixture has been recommended in patents US 3,877,450, US 3,961,609 and of nitrogen oxide, an oxygen substitute, in Patent US 3,961,609.

**[0005]** Although these solutions have achieved a partial success, they require installation of additional costly devices in engines or boilers, such as turbo chargers, oxygen tanks, measurement devices, etc. Thus, it would be more desirable to incorporate a compound directly to the fuel that could promote the complete oxidation of the fuel in the combustion chamber. These compounds would be particularly useful if they could be added simply to the fuel, fuel-oil, gasoline or diesel-oil in the corresponding tank in the form of a soluble additive.

**[0006]** For many years the use of organic peroxide derivatives has been studied as possible supplementary sources of oxygen for the fuel in the combustion chamber. For example, patent US 4,045,188 recommends as a stabiliser an additive for hydrocarbon motor fuels for internal combustion engines which consists of mixtures of ditertiarybutyl peroxide and tertiarybutyl alcohol. Certain fuel economy improvements were found at the recommended treatment levels. Patent US 4,298,351 recommends using a composition consisting of methanol and 7-25% of a tertiary alkyl peroxide. This composition can be used as a gasoline substitute as well as in mixture with conventional gasolines. The self-ignition problems in conventional gasoline engines were solved by incorporating water and isopropanol. However, the use of alcohol in these formulations can produce undesired effects such as promoting corrosion, water absorption, etc.

**[0007]** Patent EP 0255115 recommends the use of a gasoline additive composition consisting of an organic peroxide such as di-tertiary butyl peroxide, a detergent selected from among amines, diamines, polymeric amines with carboxylic acids and a suitable hydrocarbonated solvent.

**[0008]** It is also known that the performance of internal combustion engines can be improved by adding water to the liquid organic fuel. The amount of liquid fuel required to run the engine can also be reduced by adding water. A favourable consequence of the above is that the octane number of the gasoline can be increased by adding water, and a second favourable consequence is that the environmental damage resulting from combustion of organic fuel can be minimised. After over 60 years of efforts towards making full use of these advantages, this has only been achieved with high investments that sometimes have not been in relation to the benefits obtained. Thus, for example, during World War II the performance of Focker aircrafts was improved by adding water in the cylinders with a separate injector, after electrical ignition. This solution did in fact achieve an improvement of 10-15% in engine performance, but it required changing the structure of the engine and installing a special injector. For the aircraft the assembly of an additional injector and a separate water tank with the resulting increase in weight was a considerable disadvantage.

**[0009]** According to the method described in EP 0177484 A1, water is introduced in the combustion chamber as steam and not dispersed in the fuel. This requires using a special device attached to the engine.

**[0010]** These solutions have the common disadvan-

tage that they require substantial modifications of the engine and its accessory equipment, which on one hand involves significant costs and on the other the engines thus altered cannot be operated with conventional fuels. The structural changes are necessary because the fuels containing water could not be stabilised for sufficiently long periods for a safe operation, and the water/fuel mixture was made directly in the combustion chamber from its components.

**[0011]** These solutions did not achieve a widespread use because of the aforementioned structural changes required.

**[0012]** Patent US 5,156,114 claims the use of liquid fuels containing about 20 to about 80% water in internal combustion engines. This solution also requires modifying the engine, installing a catalyst in the combustion chamber to produce hydrogen from at least one part of water introduced together with the organic fuel. The hydrogen formed is burned with the organic fuel. The resulting excess power, together with the excess power resulting from the expansion of the vapour, fully compensates the loss of power due to the lower amount of organic fuel. This patent mainly addresses the use of aqueous methanol or ethanol as fuels, presenting no homogeneity problems as both of these organic compounds are completely miscible with water. In one of the examples the author also refers to the use of aqueous liquid hydrocarbonated fuels, and emphasises that the fuel must be an emulsion and that two pipes must be fitted in the engine to prevent undesirable water condensation. It describes in a general manner the type of commercially available surfactants that would be necessary to help dispersion of water in the liquid fuel, in the scope of the invention. One of the disadvantages of this solution is that the engine must be altered. Another more serious disadvantage is that when the engine is operated using a fuel that is fully miscible with water (that is, alcohol or aqueous alcohol) or with a fuel that is not miscible with water (such as a hydrocarbon or an emulsion of water in the hydrocarbon) the resulting mixture of the two types of fuel immediately causes stability problems and operational problems. Thus, when the type of fuel in the tank is not available at a service station it is necessary to use all the fuel contained in said tank, or instead to provide the vehicle with two tanks. Therefore, despite these advantages the solution disclosed in said Patent has not been used widely for combustion engines that work well with liquid hydrocarbons.

**[0013]** Finally, Patent WO 0069999 describes an additive for stabilizing water-containing fuels, consisting of an alcohol with 5 to 10 carbon atoms, 0.5 to 3 parts by weight for each part of said alcohol, an carboxylic amide with 5 to 10 carbon atoms and 3 to 10 parts by weight for each part of said alcohol of a carboxylic acid with 5 to 10 carbon atoms. The additive represents between 5-15% by weight of the liquid fuel. Although the solution disclosed in this patent allows obtaining stable emulsions of the fuel and water, the level of additivation, 5-15% by weight, required to obtain the necessary stability is relatively high and implies a considerable cost. Patent WO9944732 also describes the preparation of stable mixtures of water and oils in the presence of surfactants, although their use in combustion engines or boilers is not mentioned.

## Description of the invention

**[0014]** On our part, we have found that additives for fuels of internal combustion engines and boilers that include water, capable of improving combustion, reducing the formation of fumes, particles and carbonaceous residues and reducing the consumption of the fuel, can be obtained by a simple mixture of: a) an aqueous solution of one or more aminoacids; b) one or more alcohols containing between 1 and 5 carbon atoms; and c) reaction products of acids and basic nitrogenated compounds; and optionally d) methyl esters of fatty acids; and/or e) solvents compatible with the fuel. In the formulation of the additives it is possible also to include compounds for improving the octane and cetane numbers. The additives object of the invention are miscible with fossil fuels of petroleum origin and with bio-diesel oils (methyl or ethyl esters of fatty acids), and show no problems of instability or separation of the phases.

**[0015]** The concentration of aminoacids in the solution a) can vary greatly, generally between $1.10^{-4}$ molar and $1.10^{-1}$ molar, preferably between $1.10^{-4}$ and $1.10^{-2}$ molar. The solution can be prepared using drinking water, but it is preferable to use demineralised or distilled water. As the aminoacids can be used the aminoacids or mixtures thereof described for example in the Encyclopaedia of Chemical Processing and Design, vol. 3, p. 197-256, John J. McKetta Eds., 1977. Particularly advantageous in the scope of the present invention are histidine, phenylalanine, tryptophan and tyrosine.

**[0016]** As the alcohols b) can be used alcohols with 1 to 5 carbon atoms or their mixtures; specifically, methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 2-methyl-1-propane, 1,1-dimethylethanol, 1-pentanol, etc.

**[0017]** The ration by weight of the solution a) and the alcohols b) can vary greatly, and mainly depends on the nature of the alcohols or mixtures thereof. It is mainly chosen in order to obtain a good miscibility of the additive in the hydrocarbonated fuel employed, although these alcohols obviously improve combustion *per se* because of the oxygen they supply. In general, weight ratios a)/b) can be used of between 0.5/99.5 and 50/50, and more preferably between 2/98 and 25/75.

**[0018]** The additives object of this invention include a third component c) consisting of the reaction products of acids and basic nitrogenated products. As acids can be used acids with the general formula R-COOH, where R is an alkyl or alkenyl group with 10-36 carbon atoms, particularly fatty acids or dimers of fatty acids. As basic nitrogenated compounds can be used ammonia, hydra-

zines, urea, ethanolamine, diethanolamine, triethanolamine, primary amines such as butylamine, cyclohexylamine, hexadecylamine aniline, etc., secondary amines such as di-n-butylamine, di-n-hexylamine, etc., tertiary amines such as tri-n-butylamine, and polyamines such as diethylentriamine, triethylentriamine, tetraethylenpentamine, etc. The reaction of the acid and the amine can take place according to known methods, such as by heating the mixture of acids and amines to temperatures of 100-250°C in the presence or absence of inert solvents. The amount of component c) used is not critical and it is chosen to improve the compatibility of the components a) and b) with the fuel and to give it anticorrosive properties. Generally sufficient are amounts that, considering the amount of additive in the fuel, provide concentrations of said component c) in the fuel of between 100 and 10,000 ppm.

**[0019]** The additives object of the invention can optionally contain methyl esters of fatty acids, which enhance lubricity and provide oxygen to improve combustion. The amount of component d) is not critical.

**[0020]** Finally, the additives object of the invention can optionally contain a solvent compatible with the fuel, which is used to improve the compatibility of the other components of the additive with each other and to ensure the stability of the formulations during long periods of storage, even at very low temperatures. As a solvent e) compatible with the fuel can be used hydrocarbons or mixtures thereof, such as gasolines, diesel-oil, fuel-oil, benzene, toluene, xylenes, etc. It is also possible to use ethers such as methyl-tercbutyl ether (MTBE) and tercamyl-methyl ether (TAME), ketones such as acetone, butanone, cyclohexanone, alcohols containing 6 to 18 carbon atoms such as 2-ethylhexanol, 1-dodecanol, 1-hexadecanol, isotridecilic acid, etc. and other oxygenated solvents or mixtures thereof. The amount of solvent to use depends on the ratio of the components a) and b) and on the nature of the component b). It is recommendable to use the minimum amount of solvent e) required to achieve the desired effect of stability during storage of the additive. Larger amounts can be used but in general are not recommendable as they dilute the active components.

**[0021]** Fuels in which the additives object of the invention can be used include conventional fossil fuels derived from petroleum, such as gasolines, kerosene, diesel-oil and fuel oils. When the additives are used to improve the combustion of gasoline in internal combustion engines, other additives meant to improve the octane rating can be incorporated to the additives object of the invention, selected from among those known in the current art. When the additives are used to improve the combustion of diesel oils other additives meant to improve the cetane rating can be incorporated to the additives object of the invention, selected from among those known in the current art. The additives object of the invention can also be advantageously used to improve the combustion of biodiesel oils.

**[0022]** The most efficient concentration of the additive in the fuel to be used depends on the specific type of liquid fuel and on the desired effect regarding the reduction of pollutants and reduction of fuel consumption and on the concentration of aminoacids in the aqueous solution a). In general, one can say that the additives object of the invention are effective at levels approximately between 0.01 and 10% by weight with respect to the total weight of a) and b), and more preferably between 0.1 and 5% by weight. Any expert can easily determine the optimal levels of additive to employ in each case.

**[0023]** The invention is illustrated with the following non-limiting examples:

Example 1. *Preparation of an additive*

**[0024]** An additive according to the invention was prepared with the following formulation:

Component a): a $1.10^{-3}$ M solution of histidine in demineralised water.
Component b): isopropyl alcohol.
Weight ratio a) / b) = 5/95
Component c): reaction product of oleic acid and di-n-butylamine
Weight ratio c) / a) = 0.2/1

**[0025]** The additive was prepared simply by mixing the components a), b), c) at ambient temperature in a reactor with stirring for 5 minutes.

Example 2. *Test in gasoline engine*

**[0026]** The additive prepared according to example 1 was mixed with commercial gasoline with OR 95 in a ratio of 2 parts by volume of additive and 98 parts of gasoline. The test was performed with a single-cylinder internal combustion engine Mark III, with emissions control. Compression ratio 3:1, operation speed 400-4,000 rpm, power approximately ½ HP, cooling system: forced air, fuel injection system: injection by carburetor.

**[0027]** A dynamometer was used to initiate and load the engine. The engine speed, couple, cylinder pressure, temperature of cooling air and engine power were determined with the dynamometer. High-accuracy calibrated rotameters were used to measure the fuel and air flows.

**[0028]** Emissions of $NO_x$, total unburned hydrocarbons and CO in the exhaust gases were measured with an in-line gas analyser connected to the exhaust pipe.

**[0029]** Carbon deposits were measured gravimetrically, according to the following method: the engine was operated with a fuel-enriched air/fuel mixture, to obtain a significant formation of carbonaceous deposits on the piston after 30 minutes of operation. After each test run the engine was fully dismantled and the carbon deposited on the piston was carefully scraped and weighed with precision analytical scales. The engine was subsequently reassembled to continue the tests.

**[0030]** A commercial gasoline with OR 95 was first tested in the absence of an additive to allow a subsequent comparison. Then the same gasoline was tested with 2% by volume of an additive according to the example 1. These tests were repeated three times in order to evaluate the repeatability of the results.

**[0031]** Emissions in the exhaust gases were determined as a function of engine speed at 1500, 1750, 2000, 2250 and 2500 rpm, as well as for various equivalence ratios for the air/fuel mixture (ER defined as the actual air/fuel ratio and the stoichiometric air/fuel ratio).

**[0032]** The results obtained were as follows: emissions of CO and $NO_x$ in exhaust gases greatly depend on the air/fuel mixture equivalence ratio (ER), while those of unburned hydrocarbons (HC) do not. Emissions of HC are a minimum for an ER of 0.96-1.0, depending on the engine speed. Concentration of CO increases uniformly with lower ER's, as was expected, at constant rpm. Emissions of $NO_x$ increase with the ER.

**[0033]** The average comparative values obtained with the 95 OR gasoline with and without additive were: reduction of up to 30% of emissions of unburned hydrocarbons (HC) and carbon monoxide (CO); increased engine power with the corresponding reduced consumption above 5%; deposition of carbon reduction by 35% when using the additivated gasoline according to the invention, compared to the amount of carbon deposited with the gasoline without the additive. This use results the shows clearly increased efficiency of combustion due to the additive object of the invention.

Example 3. *Tests in industrial boilers*

**[0034]** Tests carried out in high power industrial boilers using diesel oils and fuel oils with 1-2% of the additive prepared according to example 1 resulted in fuel savings of 7-13%, depending on the level of additivation and the load demanded of the boiler.

Example 4. *Tests in domestic use boilers*

**[0035]** Tests carried out in domestic boilers for heating and hot water supply of large numbers of joint households, hotels and hospitals using diesel oil C with 2% by volume of the additive prepared according to example 1 resulted in average fuel savings of 10-15% and a reduction in emissions of carbon monoxide of approximately 50%.

Example 5. *Tests in Diesel oil automobiles*

**[0036]** Tests carried out in private-use Diesel oil automobiles using diesel oil with 2% of the additive prepared according to example 1 resulted in average fuel savings (diesel oil A) of 10-15%.

Example 5. *Tests in Diesel oil trucks*

**[0037]** Tests carried out in Diesel oil trucks of transportation fleets using diesel oil A with 2% by volume of the additive prepared according to example 1 provides average fuel savings of 10%, reducing emissions of smoke and soot by 35-40%, emissions of carbon monoxide by 50-75% and emissions of polynuclear hydrocarbons by 35%.

## Claims

1. An additive for internal combustion engine and boiler fuels consisting of mixtures of: a) aqueous solutions of one or more amino-acid selected among histidine, phenylalanine, tryptophan and tyrosine or combinations thereof; b) one or more alcohols having between 1 and 5 carbon atoms; c) reaction products of acids of formula R-COOH where R is an alkyl or alkenyl group with 10-36 carbon atoms and basic nitrogenated compounds.

2. An additive for internal combustion engine and boiler fuels consisting of mixtures of a) aqueous solutions of one or more amino-acids selected among histidine, phenylalanine, tryptophan and tyrosine or combination thereof; b) one or more alcohols having between 1 and 5 carbon atoms; c) reaction products of acids of formula R-COOH where R is an alkyl or alkenyl group with 10-36 carbon atoms and basic nitrogenated compounds, wherein, the mixture further contains: d) methyl esters of fatty acids and/or e) solvents compatible with the fuel.

3. An additive according to claim 1, **characterized in that** the concentration of amino-acids in the aqueous solutions a) is between $1.10^{-5}$ and $1.10^{-1}$ molar, preferably between $1.10^{-4}$ molar and $1.10^{-2}$ molar.

4. An additive according to claim 1, **characterized in that** the alcohol b) is isopropanol.

5. An additive according to claim 1, **characterized in that** the ratio by weight of a)/b) is between 0.5/99.5 and 50/50, and more preferably between 2/98 and 25/75.

6. An additive according to claim 1, **characterized in that** the component c) is the reaction product of fatty acids and secondary amines.

7. An additive according to claim 2, **characterized in that** the solvent e) is a hydrocarbonated solvent, an ether, a ketone, a $C_6$-$C_{18}$ alcohol or combinations thereof.

**Patentansprüche**

1. Additiv für Kraftstoffe für Verbrennungsmotoren und Heizkessel, bestehend aus Gemischen von: a) wässrigen Lösungen von einer oder mehreren Aminosäuren ausgewählt aus Histidin, Phenylalanin, Tryptophan und Tyrosin oder Kombinationen davon; b) einem oder mehreren Alkoholen, die zwischen 1 und 5 Kohlenstoffatome haben; c) Reaktionsprodukten von Säuren der Formel R-COOH, wobei R ein Alkyl- oder Alkenylrest mit 10 bis 36 Kohlenstoffatomen ist, und basischen stickstoffhaltigen Verbindungen.

2. Additiv für Kraftstoffe für Verbrennungsmotoren und Heizkessel, bestehend aus Gemischen von: a) wässrigen Lösungen von einer oder mehreren Aminosäuren ausgewählt aus Histidin, Phenylalanin, Tryptophan und Tyrosin oder Kombinationen davon; b) einem oder mehreren Alkoholen, die zwischen 1 und 5 Kohlenstoffatome haben; c) Reaktionsprodukten von Säuren der Formel R-COOH, wobei R ein Alkyl- oder Alkenylrest mit 10 bis 36 Kohlenstoffatomen ist, und basischen stickstoffhaltigen Verbindungen, wobei das Gemisch ferner enthält: d) Methylester von Fettsäuren und/oder e) mit dem Kraftstoff verträgliche Lösungsmittel.

3. Additiv nach Anspruch 1, **dadurch gekennzeichnet, dass** die Konzentration von Aminosäuren in den wässrigen Lösungen a) zwischen $1\text{-}10^{-5}$ und $1\text{-}10^{-1}$ Molar, vorzugsweise zwischen $1.10^{-4}$ Molar und $1.10^{-2}$ Molar ist.

4. Additiv nach Anspruch 1, **dadurch gekennzeichnet, dass** der Alkohol b) Isopropanol ist.

5. Additiv nach Anspruch 1, **dadurch gekennzeichnet, dass** das Gewichtsverhältnis von a)/b) zwischen 0,5/99,5 und 50/50, und stärker bevorzugt zwischen 2/98 und 25/75 ist.

6. Additiv nach Anspruch 1, **dadurch gekennzeichnet, dass** die Komponente c) das Reaktionsprodukt von Fettsäuren und sekundären Aminen ist.

7. Additiv nach Anspruch 2, **dadurch gekennzeichnet, dass** das Lösungsmittel e) ein kohlenwasserstoffhaltiges Lösungsmittel, ein Ether, ein Keton, ein $C_6$-$C_{18}$-Alkohol ist oder Kombinationen davon.

**Revendications**

1. Additif pour combustibles de moteurs à combustion interne et de chaudières, constitué d'un mélange de : a) solutions aqueuses d'un ou plusieurs aminoacides choisis parmi l'histidine, la phénylalanine, le tryptophane et la tyrosine ou des combinaisons de ceux-ci ; b) un ou plusieurs alcools ayant entre 1 et 5 atomes de carbone ; c) produits de réaction d'acides de formule R-COOH dans laquelle R est un groupe alkyle ou alcényle ayant de 10 à 36 atomes de carbone et de composés azotés basiques.

2. Additif pour combustibles de moteurs à combustion interne et de chaudières, constitué d'un mélange de : a) des solutions aqueuses d'un ou plusieurs aminoacides choisis parmi l'histidine, la phénylalanine, le tryptophane et la tyrosine ou des combinaisons de ceux-ci ; b) un ou plusieurs alcools ayant entre 1 et 5 atomes de carbone ; c) produits de réaction d'acides de formule R-COOH dans laquelle R est un groupe alkyle ou alcényle ayant de 10 à 36 atomes de carbone et de composés azotés basiques, dans lequel le mélange contient en outre : d) des esters méthyliques d'acides gras et/ou e) des solvants compatibles avec le combustible.

3. Additif selon la revendication 1, **caractérisé en ce que** la concentration d'aminoacides dans les solutions aqueuses a) est comprise entre $1.10^{-5}$ et $1.10^{-1}$ en moles, de préférence entre $1.10^{-4}$ et $1.10^{-2}$ en moles.

4. Additif selon la revendication 1, **caractérisé en ce que** l'alcool b) est de l'isopropanol.

5. Additif selon la revendication 1, **caractérisé en ce que** le rapport en poids de a)/b) est compris entre 0,5/99,5 et 50/50 et de façon plus préférée entre 2/98 et 25/75.

6. Additif selon la revendication 1, **caractérisé en ce que** le composant c) est le produit de réaction d'acides gras et d'amines secondaires.

7. Additif selon la revendication 2, **caractérisé en ce que** le solvant e) est un solvant hydrocarboné, un éther, une cétone, un alcool en $C_6$ à $C_{18}$ ou des combinaisons de ceux-ci.

## REFERENCES CITED IN THE DESCRIPTION

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