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(54) **THE USE OF 2,5-FURANDICARBOXYLIC ACID ESTERS AS LUBRICANTS**

(57) The presently claimed invention is directed to the use of novel 2,5-furandicarboxylic acid ester as lubricants. The 2,5-furandicarboxylic acid ester are obtained by reacting 2,5-furandicarboxylic acid with at least

one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol of the general formula R-OH. The present invention is also directed to a preparation process for the 2,5-furandicarboxylic acid esters.

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Description

[0001] The presently claimed invention is directed to the use of 2,5-furandicarboxylic acid esters as lubricants and a process for their preparation. The 2,5-furandicarboxylic acid esters are preferably obtained by reacting 2,5-furandicarboxylic acid with at least one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol.

[0002] The commercially available lubricant compositions are produced from a multitude of different natural or synthetic components. To improve the required properties, according to the field of use, further additives are usually added.

[0003] The various lubricants must satisfy extremely high criteria such as high viscosity index, good rheological performance, particularly at extreme temperatures, high oxidation stability, good thermal and hydrolytic stability and comparable properties.

[0004] Accordingly, high-performance lubricant oil formulations exhibit a special performance profile with respect to shear stability, low-temperature viscosity, long service life, evaporation loss, fuel efficiency, hydrolytic stability, seal compatibility and wear protection.

[0005] Such oils are currently being formulated preferentially with PAO (especially PAO 6) or group I, II or Group III mineral oils as carrier fluids, and with specific polymers like polyisobutylenes (PIBs), olefin copolymers (ethylene/propylene copolymers; OCPs), polyalkyl methacrylates (PMAs) as thickeners or viscosity index improvers in addition to the customary additive components.

[0006] Together with PAOs, low-viscosity esters are typically used in lubricant compositions, for example esters like DIDA (diisodecyl adipate), DITA (diisotridecyl adipate) or TMTC (trimethylolpropane caprylate), especially as solubilizers for polar additive types. The common esters are available by known preparation methods, and preferably from the reaction of an acid with an alcohol.

[0007] Although a wide variety of different carboxylic acid esters were developed for their use in lubricants, there is still a need for novel carboxylic acid esters which have an optimized viscosity profile over a broad temperature range, particularly at low temperatures, as well as low pour points, high viscosity index, good hydrolytic and oxidation stability high seal compatibility and favourable traction profile.

[0008] Over the previous decades environmental awareness has developed in many technical fields including the field of lubricant compositions. Accordingly, base oils of natural origin have found broad application in lubricant compositions thereby complementing the usual synthetic or mineral oils and fluids.

[0009] It still appears to be difficult to completely eliminate synthetic components of non-renewable origin in lubricating compositions due to the large industrial supply of synthetic oils and similar fluids from the global fuel producing and fuel refining industry, there is nonetheless growing interest in the industry to return to lubricating components at least partially or even completely derived from renewable sources.

[0010] Accordingly, there is the additional objective to increase the carbon content of renewable origin in lubricating compositions.

[0011] It is another object of the present invention to provide thermally and hydrolytically stable esters that can be used for the preparation of lubricants with favourable viscosity profiles over a broad temperature range including low temperatures.

[0012] The above objectives were met by preparing 2,5-furandicarboxylic acid esters obtained by reacting a mixture of branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohols with 2,5-furandicarboxylic acid.

[0013] Thus, the present invention is directed to the use of a 2,5-furandicarboxylic acid ester obtainable by reacting a mixture comprising

a) 2,5-furandicarboxylic acid, and

b) at least one alcohol of the general formula R-OH, wherein R represents a branched or linear, substituted or unsubstituted, preferably un-substituted, aliphatic C6 to C20 radical,

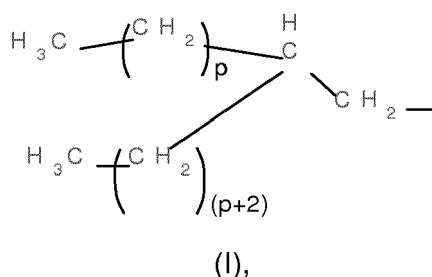
as a lubricant.

[0014] "Aliphatic C6 to C20 radical" means a saturated or unsaturated, preferably saturated, nonaromatic hydrocarbon moiety having the specified number of carbon atoms (e.g. having 6 to 20 carbon atoms, i.e. 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19 or 20 carbon atoms). In the sense of the presently claimed invention, "aliphatic C6 to C20 radicals" also include radicals wherein up to three, preferably 1 or 2, carbon atoms in the straight- or branched-chain have been replaced with a heteroatom independently selected from NH, O or S.

[0015] In connection with "aliphatic C6 to C20 radical", the term "substituted" within the scope of this invention is to be understood as meaning the substitution of hydrogen by 1, 2, 3, 4 or 5 substituents selected from the group consisting of F, Cl, Br, I, CN, NH₂, NH-C₁₋₆-alkyl, NH-C₁₋₆-alkylene-OH, N(C₁₋₆-alkyl)₂, N(C₁₋₆-alkylene-OH)₂, NO₂, SH, S-C₁₋₆-alkyl, S-benzyl, O-C₁₋₆-alkyl, O-C₁₋₆-alkylene-OH, =O, O-benzyl, C(=O)C₁₋₆-alkyl, CO₂H, CO₂-C₁₋₆-alkyl, phenyl or benzyl. The substitution of hydrogen occurs either on different atoms or on the same atom or at different positions. Polysubstitution can be carried out with the same or with different substituents.

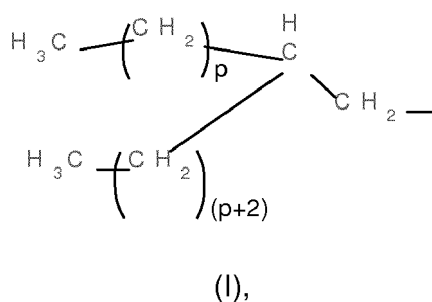
[0016] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that R is selected from the group consisting of hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, 2-ethylhexyl, 2-propylheptyl, 2-butyloctyl, 2-pentylononyl, 2-hexyldecyl, isohexyl, isoheptyl, isooctyl, isononyl, isodecyl, isoundecyl, isododecyl, isotridecyl, isotetradecyl, isopentadecyl, isohexadecyl, isoheptadecyl, isooctadecyl and mixtures thereof.

[0017] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that R is a mixture of different radicals in which at least 65 mole percent of the radicals have the general formula (I),



wherein p is 0, 1, 2, 3 or 4,

[0018] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that at least 80 mole percent of the radicals R have the general formula (I),



wherein p is 0, 1, 2, 3 or 4.

[0019] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that p is 0, 1 or 2.

[0020] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that the 2,5-furandicarboxylic acid and/or the at least one alcohol of the general formula R-OH is at least partially derived from a renewable source.

[0021] In another preferred embodiment of the present invention, the claimed use of the 2,5-furandicarboxylic acid ester is further characterized in that at least 40 mole percent of the 2,5-furandicarboxylic acid based on the total amount of 2,5-furandicarboxylic acid in the mixture are derived from a renewable source and/or at least 40 mole percent of the at least one alcohol of the general formula R-OH based on the total amount of the at least one alcohol of the general formula R-OH in the mixture are derived from a renewable source.

[0022] The present invention is also directed to a lubricant composition comprising the claimed 2,5-furandicarboxylic acid ester as defined above.

[0023] In another preferred embodiment of the present invention, the claimed lubricant composition further includes a base oil component and an additive.

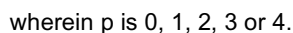
[0024] In another preferred embodiment of the present invention, the claimed lubricant composition comprises the following components:

- 5.0 to 25.0 wt.-% of the 2,5-furandicarboxylic acid ester as defined above,
- 40.0 to 90.0 wt.-% of a base oil component,
- 0.1 to 20.0 wt.-% additives.

based on the total weight of the lubricant composition.

[0025] The present invention is also directed to a process for preparing the 2,5-furandicarboxylic acid ester as defined

- [0026]** In another preferred embodiment of the present invention, the claimed process for preparing the 2,5-furandi-carboxylic acid ester is further characterized in that R is a mixture of different radicals in which at least 80 mole percent of the radicals have the general formula (I),



- [0028]** The present invention is further directed to the use of the lubricant composition as defined above in an automatic transmission fluid, a manual transmission fluid, a hydraulic fluid, a grease, a gear fluid, a metal-working fluid, a crankcase engine oil or shock absorber fluid.

- [0030]** 2,5-Furandicarboxylic acid ((FDCA, CAS Nr. 3238-40-2) is commercially available or can be prepared by processes known in the literature. Alternatively or additionally, 2,5-furandicarboxylic acid can be obtained from natural, preferably renewable, sources.

- [0032]** Renewable sources for obtaining 2,5-furandicarboxylic acid preferably include various vegetable sources, which are used for the production of fructose and glucose or similar carbohydrates. Preferably fructose and/or glucose syrups and/or non-edible carbohydrates such as (hemi)-cellulose-based carbohydrates can be used as starting materials.

- [0034]** 2,5-Furandicarboxylic acid of synthetic, non-renewable origin refers to 2,5-furandicarboxylic acid that has been

prepared by chemical technology starting from a material and/or compound which is not of renewable origin.

[0035] 2,5-Furandicarboxylic acid of renewable origin refers to 2,5-furandicarboxylic acid that has been obtained from material and/or a compound obtained from a vegetable source, preferably from agricultural production of a certain plant in which 2,5-furandicarboxylic acid is directly formed during the normal growth cycle of this plant and/or in which the normal growth cycle of said plant leads to the formation of a plant product, like preferably glucose and/or fructose or further products derived therefrom like glucose syrup, that can be subsequently processed by chemical technology or biotechnological transformation into 2,5-furandicarboxylic acid.

[0036] The plant in which the 2,5-furandicarboxylic acid or the material to be processed into 2,5-furandicarboxylic acid is formed can be a genetically modified (genetically engineered by recombinant DNA or RNA technology) plant. Alternatively or additionally, the 2,5-furandicarboxylic acid is obtained starting from material obtained from a natural, wild-type plant which has not been modified by recombinant technology.

[0037] When providing the 2,5-furandicarboxylic acid for obtaining the 2,5-furandicarboxylic acid ester used as lubricant in the present invention, at least 40 mole percent of the 2,5-furandicarboxylic acid based on the total amount of 2,5-furandicarboxylic acid used in the preparation of the 2,5-furandicarboxylic acid ester are preferably derived from a renewable source or origin. More preferably, at least 50 mole percent, or more preferably at least 65 mole percent, even more preferably at least 75 mole percent, and most preferably at least 85 mole percent of the 2,5-furandicarboxylic acid used for obtaining the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention can be derived from a renewable source in the claimed preparation process.

[0038] However, in another very preferred embodiment of the present invention, the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention is obtained from 2,5-furandicarboxylic acid which is fully of renewable origin, i.e. the 2,5-furandicarboxylic acid consists of at least 90 mole percent, at least 95 mole percent, or even 100 mole percent of material from a renewable source.

[0039] Regarding the grade of chemical purity, 2,5-furandicarboxylic acid as used in the present invention is either substantially pure, i.e. includes at least 80 percent by weight, preferably at least 90 percent by weight, more preferably at least 95 percent by weight, even more preferably at least 97 percent by weight, even more preferably at least 98.0 percent by weight, or most preferably at least 99.0 percent by weight, or even at least 99.5 percent by weight of 2,5-furandicarboxylic acid, based on the total amount of the corresponding sample of 2,5-furandicarboxylic acid.

[0040] In addition, 2,5-furandicarboxylic acid as used in the present invention preferably has very low level of unsaturated components. The content of unsaturated components is normally less than 5 percent by weight, preferably less than 3 percent by weight, more preferably less than 1 percent by weight, even more preferably less than 0.5 percent by weight, and most preferably less than 0.1 percent by weight, based on the total weight of 2,5-furandicarboxylic acid.

[0041] Most generically, the at least one alcohol of the general formula R-OH to be used in the preparation of the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention is defined by a radical R representing a branched or linear, substituted or unsubstituted aliphatic hydrocarbon moiety having from 6 to 20 carbon atoms.

[0042] Most preferably, the radical R denotes a moiety selected from the group consisting of hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, 2-ethylhexyl, 2-propylheptyl, 2-butyloctyl, 2-pentylononyl, 2-hexyldecyl, isohexyl, isoheptyl, isooctyl, isononyl, isodecyl, isoundecyl, isododecyl, isotridecyl, isotetradecyl, isopentadecyl, isohexadecyl, isoheptadecyl, isooctadecyl and mixtures thereof.

[0043] Even more preferably, the radical R denotes a branched or linear, substituted or unsubstituted aliphatic hydrocarbon moiety having from 8 to 18 carbon atoms, or even more preferably, 8 to 16 carbon atoms.

[0044] Especially preferred moieties having 8 to 18 carbon atoms include octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, 2-ethylhexyl, 2-propylheptyl, 2-butyloctyl, 2-pentylononyl, 2-hexyldecyl, isohexyl, isoheptyl, isooctyl, isononyl, isodecyl, isoundecyl, isododecyl, isotridecyl, isotetradecyl, isopentadecyl, isohexadecyl, isoheptadecyl, isooctadecyl and mixtures thereof.

[0045] Especially preferred moieties having 8 to 16 carbon atoms include octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, 2-ethylhexyl, 2-propylheptyl, 2-butyloctyl, 2-pentylononyl, 2-hexyldecyl, isohexyl, isoheptyl, isooctyl, isononyl, isodecyl, isoundecyl, isododecyl, isotridecyl, isotetradecyl, isopentadecyl, isohexadecyl, and mixtures thereof.

[0046] The alcohol with the radical R being 2-propylheptyl, i.e. 2-propylheptanol, is commercially available from BASF SE, Ludwigshafen and represents a mixture of 93.0 wt.-% 2-propyl-heptanol, 2.9 wt.-% 2-propyl-4-methyl-hexanol, 3.9 wt.-% 2-propyl-5-methylhexanol and 0.2 wt.-% 2-isopropylheptanol.

[0047] When providing the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol for obtaining the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention, at least 40 mole percent of the at least one alcohol of the general formula R-OH based on the total amount of the at least one alcohol of the general formula R-OH used in the preparation of the 2,5-furandicarboxylic acid ester are preferably derived from a renewable source or origin. Preferably, at least 50 mole percent, more preferably at least 65 mole percent, even more preferably at least 75 mole percent, and most preferably at least 85 mole percent of the at least one alcohol of the general formula R-OH used for obtaining the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention

can be derived from a renewable source in the claimed preparation process. The term at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol "of renewable origin" is defined in a similar manner like 2,5-furandicarboxylic acid of renewable origin is defined above. Accordingly, an alcohol of renewable origin is one which is prepared, isolated or derived starting from a natural, renewable source or material obtained for instance from a plant.

[0048] In one preferred embodiment of the present invention, the 2,5-furandicarboxylic acid ester is obtained from 2,5-furandicarboxylic acid of renewable origin, exclusively, and the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol, especially a mixture of Guerbet alcohols as defined herein, which also is of renewable origin, exclusively. Accordingly, it is preferred that the 2,5-furandicarboxylic acid ester is formed exclusively from components of renewable origin.

[0049] Alternatively, the 2,5-furandicarboxylic acid and/or the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol are only partially derived from a renewable source thereby leading to a 2,5-furandicarboxylic acid ester which is also only of partial renewable origin based on the combined amounts of the respective partial amount of 2,5-furandicarboxylic acid of renewable origin and the respective partial amount of the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol, especially a mixture of Guerbet alcohols as defined herein, of renewable origin.

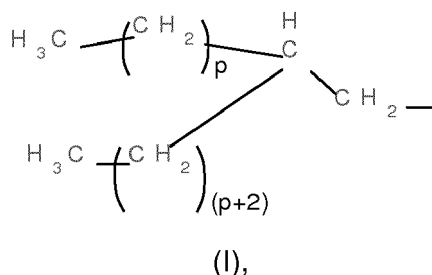
[0050] One preferred alcohol of the general formula R-OH, or mixture of alcohols R-OH, which is used in the present invention for preparing the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention, is a so-called "Guerbet alcohol".

[0051] The term "Guerbet alcohol" is known to those skilled in the art. Reference is made by way of example to "Alcohols, Aliphatic", page 5 and page 10 respectively in Ullmann's Encyclopedia of Industrial Chemistry, Seventh Ed., Electronic Release, 2008, Wiley-VCH, Weinheim, N.Y., and the literature cited there.

[0052] The term "Guerbet alcohol" as used in the present invention relates to alcohols, or a mixture of alcohols, which is obtained by the so-called Guerbet reaction. Accordingly, the mixture of alcohols as used in the present invention for obtaining the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention is a mixture of alcohols obtained from the Guerbet reaction, particularly as defined in the above references.

[0053] In the course of the Guerbet reaction, primary alcohols are ultimately dimerized to α -branched primary alcohols in the presence of suitable catalysts. According to the literature, the primary products formed from the alcohols are aldehydes which subsequently dimerize to saturated alcohols by aldol condensation with elimination of water and subsequent hydrogenation. As well as the main product, different by-products can also form, for example unsaturated α -branched primary alcohols if the hydrogenation of the double bond does not proceed to completion, or more particularly α -branched primary alcohols which have additional branches in the side chain or main chain.

[0054] In another embodiment, one preferred R radical of the at least one branched or linear, substituted or unsubstituted aliphatic alcohol of the general formula R-OH is a radical derived from a Guerbet alcohol, i.e. obtained from the Guerbet reaction comprising or containing a mixture of different radicals in which at least 50 mole %, or 65 mole percent, more preferably at least 70 mole percent, even more preferably at least 80 mole %, and most preferably at least 90 mole % of the radicals based on the total amount of radicals in the mixture have the general formula (I).



wherein p is 0, 1, 2, 3 or 4; preferably 0,1 or 2, and more preferably 2;

[0055] The invention further relates to the use of the claimed 2,5-furandicarboxylic acid esters as additives, co-solvents or base oils in lubricant compositions and fuel additives.

[0056] By the terms "lubricant" or "lubricant composition", as used in the presently claimed invention, is meant a substance or composition capable of reducing friction between moving surfaces.

[0057] The lubricant compositions according to the present invention can comprise the 2,5-furandicarboxylic acid esters as one important component, for instance in a rather limited amount of from 0.1 to 50.0 percent by weight, preferably 3.0 to 40.0 percent by weight, more preferably 5.0 to 30.0 percent by weight, or even more preferably 10.0 to 25.0 percent by weight, or, alternatively, as main component in the lubricant composition of the present invention, in an amount of from 50.0 percent by weight to 100.0 percent by weight, preferably 60.0 percent by weight to 95.0 percent by weight, more preferably 65.0 percent by weight to 90.0 percent by weight, or even more preferably 75.0 percent by

weight to 85.0 percent by weight, based on the total amount of lubricant composition.

[0058] In another preferred embodiment of the present invention, the 2,5-furandicarboxylic acid esters are preferably used in an amount of 3.5 to 45 percent by weight, more preferably in an amount of from 5.0 to 35.0 percent by weight, and most preferably in an amount of 10.0 to 30.0 percent by weight, based on the total amount of the lubricant composition.

[0059] One preferred lubricant composition of the present invention includes the following components:

- 5.0 to 25.0 wt.-% of 2,5-furandicarboxylic acid ester as defined above,
- 40.0 to 90.0 wt.-% of a base oil component,
- 0.1 to 20.0 wt.-% additives,

based on the total weight of the lubricant composition, wherein, preferably, the additives can include viscosity index improvers, like PMAs, OCPs and/or PIBs, in an amount of 10 to 30 wt.-%.

[0060] The following further lubricant compositions comprising the 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention are also preferred (all numbers in percent by weight); the second range given ("or") is an even more preferred range, respectively:

	Lubricant type A	Lubricant type B	Lubricant type C
Base oil	50.0 to 85.0; or 65.0 to 75.0;	25.0 to 75.0; or 35.0 to 65.0;	0 to 45.0; or 5.0 to 20.0;
2,5-furandicarboxylic acid ester	5.0 to 20.0; or 10.0 to 17.0;	20.0 to 50.0; or 30.0 to 45.0;	50.0 to 85.0; or 60.0 to 80.0;
Additives	10.0 to 30.0; or 15.0 to 25.0;	5.0 to 25.0; or 10.0 to 20.0;	5.0 to 15.0; or 8.0 to 13.0;

[0061] The following lubricant compositions are especially preferred (all numbers in percent by weight); the second range ("or") given is an even more preferred range, respectively:

	Lubricant type A'	Lubricant type B'	Lubricant type C'
Base oil	40.0 to 78.0; or 45.0 to 70.0;	25.0 to 75.0; or 32.0 to 60.0;	0 to 45.0; or 20.0 to 30.0;
2,5-furandicarboxylic acid ester	5.0 to 15.0; or 8.0 to 15.0;	20.0 to 50.0; or 30.0 to 40.0;	50.0 to 85.0; or 65.0 to 80.0;
Additives (without VI improvers)	5.0 to 15.0; or 8.0 to 15.0;	0 to 10.0; or 5.0 to 8.0;	0.0 to 5.0; or 2.0 to 4.0;
Additives (VI improvers only)	5.0 to 30.0; or 10.0 to 28.0;	5.0 to 15.0; or 8.0 to 12.0;	5.0 to 10.0; or 6.0 to 8.0;

[0062] Viscosity index (VI) improvers are one possible class of additives that can be optionally used in the lubricant compositions of the present invention. Examples of viscosity index improvers include for instance OCPs, PMAs and/or PIBs, and are also disclosed below where the additives are listed in more detail (see point 5).

[0063] The lubricant compositions according to the present invention may further include base oils or co-solvents.

[0064] Preferred base oils contemplated for optional use in the lubricating oil compositions according to the present invention include mineral oils, poly-alpha-olefin synthetic oils and mixtures thereof.

[0065] Suitable base oils also include base stocks obtained by isomerization of synthetic wax and slack wax, as well as base stocks produced by hydrocracking (rather than solvent extracting) the aromatic and polar components of the crude. In general, both the mineral and synthetic base oils will each have a kinematic viscosity ranging from about 1 to about 40 cSt at 100°C, although typical applications will require each oil to have a viscosity ranging from about 1 to about 10 cSt at 100°C.

[0066] The mineral oils useful as optional components in the lubricant compositions according to the present invention include all common mineral oil base stocks. This includes oils that are naphthenic, paraffinic or aromatic in chemical structure.

[0067] Naphthenic oils are made up of methylene groups arranged in ring formation with paraffinic side chains attached to the rings. The pour point is generally lower than the pour point for paraffinic oils.

[0068] Paraffinic oils comprise saturated, straight chain or branched hydrocarbons.

[0069] The straight chain paraffins of high molecular weight raise the pour point of oils and are often removed by dewaxing.

[0070] Aromatic oils are hydrocarbons of closed carbon rings of a semi-unsaturated character and may have attached side chains. This oil is more easily degraded than paraffinic and naphthalenic oils leading to corrosive by-products.

[0071] In reality, a base stock will normally contain a chemical composition which contains some proportion of all three oils (paraffinic, naphthenic and aromatic). For a discussion of types of base stocks, see Motor Oils and Engine Lubrication by A. Schilling, Scientific Publications, 1968, section 2.2 through 2.5.

[0072] Further optional base oils include gas to liquid oils (GTL). Gaseous sources include a wide variety of materials such as natural gas, methane, C1-C3 alkanes, landfill gases, and the like. Such gases may be converted to liquid hydrocarbon products suitable for use as lubricant base oils by a gas to liquid (GTL) process, such as the process described in U.S. patent 6,497,812, of which the disclosure is incorporated herein by reference.

[0073] Base oils derived from a gaseous source, hereinafter referred to as "GTL base oils", typically have a viscosity index of greater than about 130, a sulfur content of less than about 0.3 percent by weight, contain greater than about 90 percent by weight saturated hydrocarbons (isoparaffins), typically from about 95 to about 100 percent by weight branched aliphatic hydrocarbons, have a pour point of below -15 to -20°C.

[0074] The GTL base oils may be mixed with more conventional base oils such as Groups I to V as specified by API. For example, the base oil component of the lubricant compositions of the present invention may include 1 to 100 percent by weight of GTL base oil.

[0075] Thus, the lubricating oil composition of the present invention may optionally be at least partially derived from a gaseous source.

[0076] Oils may be refined by conventional methodology using acid, alkali, and clay or other agents such as aluminum chloride, or they may be extracted oils produced, for example, by solvent extraction with solvents such as phenol, sulfur dioxide, furfural, dichloroethyl ether, etc.

[0077] They may be hydro-treated or hydro-refined, dewaxed by chilling or catalytic dewaxing processes, or hydro-cracked. The mineral oil may be produced from natural crude sources or be composed of isomerized wax materials or residues of other refining processes. The preferred synthetic oils are oligomers of α -olefins, particularly oligomers of 1-decene, also known as polyalphaolefins or PAO's.

[0078] The base oils may be derived from refined, re-refined oils, or mixtures thereof. Unrefined oils are obtained directly from a natural source or synthetic source (e.g., coal, shale, or tar sands bitumen) without further purification or treatment. Examples of unrefined oils include a shale oil obtained directly from a retorting operation, petroleum oil obtained directly from distillation, or ester oil obtained directly from an esterification process, each of which is then used without further treatment. Refined oils are similar to the unrefined oils except that refined oils have been treated in one or more purification steps to improve one or more properties. Suitable purification techniques include distillation, hydro-treating, dewaxing, solvent extraction, acid or base extraction, filtration, and percolation, all of which are known to those skilled in the art. Re-refined oils are obtained by treating used oils in processes similar to those used to obtain the refined oils. These re-refined oils are also known as reclaimed or reprocessed oils and are often additionally processed by techniques for removal of spent additives and oils breakdown products.

[0079] In the lubricant compositions according to the present invention comprising the 2,5-furandicarboxylic acid ester of the present invention, it is optional to include other esters being capable of reducing friction between moving surfaces.

[0080] For instance, the lubricant compositions of the present invention can further comprise other monocarboxylic acid esters or dicarboxylic acid esters. Both additional types of optional esters are suitable for reducing friction and can be added together or individually to the lubricant compositions of the present invention.

[0081] The monocarboxylic and dicarboxylic acid esters that can be optionally used in addition are present in the lubricant compositions either individually, or in the form of mixtures comprising at least one monocarboxylic acid ester and at least one dicarboxylic acid ester.

[0082] Such monocarboxylic and dicarboxylic acid esters are obtained from known procedures, preferably by esterification of the corresponding monocarboxylic and/or dicarboxylic acid with the corresponding alcohol or mixture of alcohols.

[0083] Representative monocarboxylic acids include n-butanoic acid, n-pentanoic acid, n-hexanoic acid, n-heptanoic acid, n-octanoic acid, n-nonanoic acid, n-decanoic acid, isobutanoic acid, isopentanoic acid, isohexanoic acid, isohexanoic acid, isooctanoic acid, 2-ethylhexanoic acid, isononanoic acid, 3,5,5-trimethylhexanoic acid, and isodecanoic acid.

[0084] Representative dicarboxylic acid esters can be obtained from aliphatic dicarboxylic acids. In preferred modes of the present invention, the optional dicarboxylic acid esters can be obtained from dicarboxylic acids selected from the group consisting of glutaric acid, diglycolic acid, succinic acid, azelaic acid, sebacic acid, 1,4-cyclohexanedicarboxylic acid, adipic acid, 2,6-decahydronaphthalene dicarboxylic acid, 1,3-cyclohexanedicarboxylic acid, and 2,5-norbornanedicarboxylic acid. A very preferred aliphatic dicarboxylic acid is adipic acid. Instead of the acids, their anhydrides can also be used.

[0085] Representative alcohols to be used for obtaining other optional monocarboxylic acid esters and/or dicarboxylic

acid esters include 2-propylheptanol, 2-propyl-4-methyl-hexanol, 2-propyl-5-methyl-hexanol, 2-isopropyl-4-methyl-hexanol, 2-isopropyl-5-methyl-hexanol, 2-propyl-4,4-dimethylpentanol, 2-ethyl-2,4-dimethylhexanol, 2-ethyl-2-methyl-heptanol, 2-ethyl-2,5-dimethylhexanol and 2-isopropyl-heptanol.

[0086] Preferably the alcohol mixture comprises 80 to 95 percent by weight of 2-n-propyl-heptanol, 1.0 to 10 percent by weight of 2-propyl-4-methyl-hexanol, 1.0 to 10 percent by weight of 2-propyl-5-methyl-hexanol and 0.1 to 2.0 percent by weight of 2-isopropyl-heptanol, whereby the weight of each component is related to the total weight of the monoalcohols.

[0087] More preferably the mixture comprises 91.0 to 95.0 percent by weight of 2-n-propyl-heptanol, 2.0 to 5.0 percent by weight of 2-propyl-4-methyl-hexanol, 3.0 to 5.0 percent by weight of 2-propyl-5-methyl-hexanol and 0.1 to 0.8 percent by weight of 2-isopropyl-heptanol, whereby the weight of each component is related to the total weight of the monoalcohols.

[0088] In another embodiment, an additional optional carboxylic acid ester is obtained by reacting a mixture comprising adipic acid, 2-propyl-heptanol, 2-propyl-4-methyl-hexanol and 2-propyl-5-methyl-hexanol.

[0089] In another embodiment, an additional optional carboxylic acid ester to be present in the lubricant compositions of the present invention is obtained by reacting a mixture comprising adipic acid and 80 to 95 percent by weight of 2-n-propyl-heptanol, 1.0 to 10 percent by weight of 2-propyl-4-methyl-hexanol, 1.0 to 10 percent by weight of 2-propyl-5-methyl-hexanol, and 0.1 to 2.0 percent by weight of 2-isopropyl-heptanol, whereby the weight of each component is related to the total weight of the monoalcohols.

[0090] The addition of at least one additive, like an additional customary oil additive, to the lubricating oil compositions of the present invention is possible but not mandatory in every case. The mentioned lubricant compositions, e.g. greases, gear fluids, metal-working fluids and hydraulic fluids, may additionally comprise further additives that are added in order to improve their basic properties still further.

[0091] Such additives include: further antioxidants or oxidation inhibitors, corrosion inhibitors, friction modifiers, metal passivators, rust inhibitors, anti-foamants, viscosity index enhancers, additional pour-point depressants, dispersants, detergents, further extreme-pressure agents and/or anti-wear agents.

[0092] Such additives are present in the amounts customary for each of them, which range in each case from 0.01 to 30.0 percent by weight, preferably from 0.05 to 20.0 percent by weight, more preferably from 0.1 to 10.0 percent by weight, and even more preferably 0.2 to 5.0 percent by weight, based on the total weight of the lubricating oil composition. Examples of further additives are given below:

1. Examples of phenolic antioxidants:

1.1. Alkylated monophenols: 2,6-di-tert-butyl-4-methylphenol, 2-butyl-4,6-dimethylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(alpha-methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, linear nonylphenols or nonylphenols branched in the side chain, such as, for example, 2,6-dinonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)-phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)-phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)-phenol and mixtures thereof;

1.2. Alkylthiomethylphenols: 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-didodecylthiomethyl-4-nonylphenol;

1.3. Hydroquinones and alkylated hydroquinones: 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis(3,5-di-tert-butyl-4-hydroxyphenyl) adipate;

1.4. Tocopherols: alpha-, beta-, gamma- or delta-tocopherol and mixtures thereof (vitamin E);

1.5. Hydroxylated thiodiphenyl ethers: 2,2'-thio-bis(6-tert-butyl-4-methylphenol), 2,2'-thio-bis(4-octylphenol), 4,4'-thio-bis(6-tert-butyl-3-methylphenol), 4,4'-thio-bis(6-tert-butyl-2-methylphenol), 4,4'-thio-bis(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxy-phenyl)disulfide;

1.6. Alkylidene bisphenols: 2,2'-methylene-bis(6-tert-butyl-4-methylphenol), 2,2'-methylene-bis(6-tert-butyl-4-ethylphenol), 2,2'-methylene-bis[4-methyl-6-(alpha-methylcyclohexyl)phenol], 2,2'-methylene-bis(4-methyl-6-cyclohexylphenol), 2,2'-methylene-bis(6-nonyl-4-methylphenol), 2,2'-methylene-bis(4,6-di-tert-butylphenol), 2,2'-ethylidene-bis(4,6-di-tert-butylphenol), 2,2'-ethylidene-bis(6-tert-butyl-4-isobutylphenol), 2,2'-methylene-

bis[6-(alpha -methylbenzyl)-4-nonylphenol], 2,2'-methylene-bis[6-(alpha, alpha -dimethyl-benzyl)-4-nonylphenol], 4,4'-methylene-bis(2,6-di-tert-butylphenol), 4,4'-methylene-bis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)-butyrate], bis(3-tert-butyl-4-hydroxy-5-methylphenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)-propane, 2,2-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane;

1.7. O-, N- and S-benzyl compounds: 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzyl-mercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzyl-mercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzyl-mercaptoacetate;

1.8. Hydroxybenzylated malonates: dioctadecyl-2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, dioctadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)malonate, didodecyl-mercaptoethyl-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl) malonate, di[4-(1,1,3,3-tetramethylbutyl)-phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate;

1.9. Hydroxybenzyl aromatic compounds: 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol;

1.10. Triazine compounds: 2,4-bis-octylmercapto-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate;

1.11. Acylaminophenols: 4-hydroxylauric acid anilide, 4-hydroxystearic acid anilide, N-(3,5-di-tert-butyl-4-hydroxyphenyl)-carbamic acid octyl ester;

1.12. Esters of beta-(5-tert-butyl-4-hydroxy-3-methylphenyl) propionic acid: with polyhydric alcohols, e.g. with 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl) oxalic acid diamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane;

1.13. Esters of beta-(3,5-di-tert-butyl-4-hydroxyphenyl) propionic acid, gamma-(3,5-dicyclohexyl-4-hydroxyphenyl) propionic acid, 3,5-di-tert-butyl-4-hydroxyphenylacetic acid: with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, isooctanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis-hydroxyethyl oxalic acid diamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane;

1.14. Amides of beta-(3,5-di-tert-butyl-4-hydroxyphenyl) propionic acid: N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamine, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hydrazine;

1.15. Ascorbic acid (vitamin C);

1.16. Aminic antioxidants: N,N'-diisopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-di(naphth-2-yl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-

phenyl-p-phenylenediamine, 4-(p-toluenesulfonamido)-diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, 4-n-butylaminophenol, 4-butyrylamino-phenol, 4-nonanoylamino-phenol, 4-dodecanoylamino-phenol, 4-octadecanoylamino-phenol, di(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethyl phenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-di[(2-methylphenyl)amino]-ethane, 1,2-di(phenylamino)propane, (o-tolyl)biguanide, di[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, mixture of mono- and di-alkylated tert-butyl/tert-octyl-diphenylamines, mixture of mono- and di-alkylated nonylidiphenylamines, mixture of mono- and di-alkylated dodecyldiphenylamines, mixture of mono- and di-alkylated isopropyl/isohexyl-diphenylamines, mixtures of mono- and di-alkylated tert-butylidiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, mixture of mono- and di-alkylated tert-butyl/tert-octylphenothiazines, mixtures of mono- and di-alkylated tert-octylphenothiazines, N-allylphenothiazine, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylenediamine, bis(2,2,6,6-tetramethylpiperidin-4-yl)sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

2. Examples of further antioxidants: aliphatic or aromatic phosphites, esters of thiodipropionic acid or thiodiacetic acid or salts of dithiocarbamic acid, 2,2,12,12-tetramethyl-5,9-dihydroxy-3,7,11-trithiamidecane and 2,2,15,15-tetramethyl-5,12-dihydroxy-3,7,10,14-tetrathiahexadecane.

3. Examples of metal deactivators, e.g. for copper:

3.1. Benzotriazoles and derivatives thereof: 2-mercaptobenzotriazole, 2,5-dimercaptobenzotriazole, 4- or 5-alkylbenzotriazoles (e.g. tolutriazole) and derivatives thereof, 4,5,6,7-tetrahydrobenzotriazole, 5,5'-methylenebis-benzotriazole; Mannich bases of benzotriazole or tolutriazole, such as 1-[di(2-ethylhexyl)aminomethyl]tolutriazole and 1-[di(2-ethylhexyl)aminomethyl]benzotriazole; alkoxyalkylbenzotriazoles, such as 1-(nonyloxy-methyl)benzotriazole, 1-(1-butoxyethyl)-benzotriazole and 1-(1-cyclohexyloxybutyl)-tolutriazole;

3.2. 1,2,4-Triazoles and derivatives thereof: 3-alkyl-(or -aryl-) 1,2,4-triazoles, Mannich bases of 1,2,4-triazoles, such as 1-[di(2-ethylhexyl)aminomethyl]-1,2,4-triazole; alkoxyalkyl-1,2,4-triazoles, such as 1-(1-butoxyethyl)-1,2,4-triazole; acylated 3-amino-1,2,4-triazoles;

3.3. Imidazole derivatives: 4,4'-methylene-bis(2-undecyl-5-methyl) imidazole and bis[(N-methyl)imidazol-2-yl]carbinol-octyl ether;

3.4. Sulfur-containing heterocyclic compounds: 2-mercaptobenzothiazole, 2,5-dimercapto-1,3,4-thiadiazole, 2,5-dimercaptobenzothiadiazole and derivatives thereof; 3,5-bis[di(2-ethylhexyl)aminomethyl]-1,3,4-thiadiazolin-2-one;

3.5. Amino compounds: salicylidene-propylenediamine, salicylaminoguanidine and salts thereof.

4. Examples of rust inhibitors:

4.1. Organic acids, their esters, metal salts, amine salts and anhydrides: alkyl- and alkenylsuccinic acids and their partial esters with alcohols, diols or hydroxycarboxylic acids, partial amides of alkyl- and alkenyl-succinic acids, 4-nonylphenoxyacetic acid, alkoxy- and alkoxyethoxycarboxylic acids, such as dodecyloxyacetic acid, dodecyloxy (ethoxy)acetic acid and amine salts thereof, and also N-oleoyl-sarcosine, sorbitan monooleate, lead naphthenate, alkenylsuccinic acid anhydrides, e.g. dodecenylsuccinic acid anhydride, 2-(2-carboxyethyl)-1-dodecyl-3-methylglycerol and salts thereof, especially sodium and triethanolamine salts thereof.

4.2. Nitrogen-containing compounds:

4.2.1. Tertiary aliphatic or cycloaliphatic amines and amine salts of organic and inorganic acids, e.g. oil-soluble alkylammonium carboxylates, and 1-[N,N-bis(2-hydroxyethyl)amino]-3-(4-nonylphenoxy)propan-2-ol;

4.2.2. Heterocyclic compounds: substituted imidazolines and oxazolines, e.g. 2-heptadecenyl-1-(2-hydroxyethyl)-imidazoline;

4.2.3. Sulfur-containing compounds: barium dinonylnaphthalene sulfonates, calcium petroleum sulfonates, alkylthio-substituted aliphatic carboxylic acids, esters of aliphatic 2-sulfocarboxylic acids and salts thereof.

5. Examples of additional viscosity index enhancers: polyacrylates, polymethacrylates (PMAs), nitrogen containing polymethylmethacrylates, vinylpyrrolidone/methacrylate copolymers, polyvinylpyrrolidones, polybutenes, polyisobutylenes (PIBs), olefin copolymers such as ethylenepropylene copolymers (OCPs), styrene-isoprene copolymers, hydrated styrene-isoprene copolymers, styrene/acrylate copolymers and polyethers. Multifunctional viscosity improvers, which also have dispersant and/or antioxidancy properties are known and may optionally be used in addition to the products of this invention. Conventional polyisobutylenes (PIBs), like Lubrazol® 8406), and/or oligomeric copolymers (OCPs), like Lubrazol® 8407 are one preferred addition of additional viscosity index improvers in the lubricant composition of the present invention comprising 2,5-furandicarboxylic acid esters.

6. Examples of pour-point depressants: polymethacrylates, ethylene/vinyl acetate copolymers, alkyl polystyrenes, fumarate copolymers, alkylated naphthalene derivatives.

7. Examples of dispersants/surfactants: polybutenylsuccinic acid amides or imides, polybutenylphosphonic acid derivatives, basic magnesium, calcium and barium sulfonates and phenolates.

8. Examples of extreme-pressure and anti-wear additives: sulfur- and halogen-containing compounds, e.g. chlorinated paraffins, sulfurized olefins or vegetable oils (soybean oil, rape oil), alkyl- or aryl-di- or -tri-sulfides, benzotriazoles or derivatives thereof, such as bis(2-ethylhexyl)aminomethyl tolutriazoles, dithiocarbamates, such as methylene-bis-dibutyldithiocarbamate, derivatives of 2-mercaptobenzothiazole, such as 1-[N,N-bis(2-ethylhexyl)aminomethyl]-2-mercapto-1H-1,3-benzothiazole, derivatives of 2,5-dimercapto-1,3,4-thiadiazole, such as 2,5-bis(tert-nonyldithio)-1,3,4-thiadiazole.

9. Examples of coefficient of friction reducers: lard oil, oleic acid, tallow, rape oil, sulfurized fats, amides, amines. Further examples are given in EP-A-0 565 487.

10. Examples of special additives for use in water/oil metal-working fluids and hydraulic fluids: Emulsifiers: petroleum sulfonates, amines, such as polyoxyethylated fatty amines, non-ionic surface-active substances; buffers: such as alkanolamines; biocides: triazines, thiazolinones, tris-nitromethane, morpholine, sodium pyridenethiol; processing speed improvers: calcium and barium sulfonates.

[0093] Depending on the end use applications, small quantities of additives such as anti-misting agents may be also optionally added in an amount ranging from 0.05 to 5.0% by vol. in one embodiment, and less than 1 wt. %, in other embodiments.

[0094] For certain applications, a small amount of foam inhibitors in the prior art can also be added to the composition in an amount ranging from 0.02 to 15.0 wt. %.

[0095] The compositions may further comprise oil soluble metal deactivators in an amount of 0.01 to 0.5 vol. % (based on the final oil volume).

[0096] Esterification of the 2,5-furandicarboxylic acid can be carried out according to established procedures which are known to the skilled person. Additional detailed descriptions for carrying out the esterification reaction are disclosed in US 6,310,235, US 5,324,853, DE-A 2612355 (Derwent Abstract No. DW 77-72638 Y) or DE-A 1945359 (Derwent Abstract No. DW 73-27151 U). These documents and the corresponding descriptions therein are herewith incorporated by reference.

[0097] In a preferred embodiment of the preparation process of the present invention, the 2,5-furandicarboxylic acid esters to be used as lubricants in the present invention can be obtained by initially providing 2,5-furandicarboxylic acid and/or the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol from a non-renewable and/or renewable source as defined above. It is especially preferred that all components from which the ester is formed, i.e. acid and alcohol is of renewable origin.

[0098] In a first step, the 2,5-furandicarboxylic acid or a suitable derivative thereof is provided. Preferably, the 2,5-furandicarboxylic acid can be esterified with the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol of the general formula R-OH by use of the corresponding acyl halogenide, preferably the acyl chloride or acyl bromide, or the respective anhydride of 2,5-furandicarboxylic acid.

[0099] Then, in a next step, a mixture is prepared of the 2,5-furandicarboxylic acid with the at least one C6 to C20 alcohol of the general formula R-OH, wherein the radical R denotes a branched or linear, substituted or unsubstituted aliphatic hydrocarbon moiety having from 6 to 20 carbon atoms.

[0100] More preferably, the radical R denotes a branched or linear, substituted or unsubstituted aliphatic hydrocarbon

[0101] Most preferably, the radical R denotes a moiety selected from the group consisting of decyl, isodecyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, 2-propylheptyl, 2-butyloctyl, 2-pentylnonyl and 2-hexyldeceyl, as well as mixtures thereof.

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[0103] In the subsequent process stage or step, the esterification of the 2,5-furandicarboxylic acid is carried out with the mixture of the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol of the general formula R-OH as defined above. This esterification reaction of the 2,5-furandicarboxylic acid preferably includes the following optional or preferred process features.

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[0109] WO 02/038531 A1 discloses processes for the preparation of esters, comprising

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increased pressure. Preferably, the esterification is carried out at ambient pressure or decreased pressure.

[0112] The esterification of the present invention can be carried out in the absence of an additional solvent or in the presence of an organic solvent, preferably an organic solvent which is chemically inert under the esterification conditions. Examples for organic solvents include aliphatic hydrocarbons, halogenated aliphatic hydrocarbons, aromatic and substituted aromatic hydrocarbons or ethers. Very preferred solvents are selected from pentane, hexane, heptane, ligroin, petroleum ether (benzene), cyclohexane, dichloromethane, trichloromethane, tetrachloromethane, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, dibutylether, tetrahydrofuran, dioxane and mixtures thereof.

[0113] Esterification can be carried out in the absence or presence of an inert gas. The term "inert gas" refers to gas which does not react with the educts, reagents, solvents or the products formed in the reaction under the given process conditions. Preferably, esterification is carried out without the addition of an inert gas.

[0114] Optionally, the 2,5-furandicarboxylic acid ester which was thus obtained can be further purified by drying and filtering.

[0115] In another preferred embodiment, the reaction between the 2,5-furandicarboxylic acid and the mixture of the at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol of the general formula R-OH can be preferably carried out using stoichiometric amounts of 2,5-furandicarboxylic acid and alcohol based on the number of reactive OH- and CO₂H-groups, particularly when entrainers are used.

[0116] However, one preference can be optionally given to using a stoichiometric excess (based on the number of reactive OH- and CO₂H-groups) of the alcohol component of from 0.05 to 1.0 mole per mole of 2,5-furandicarboxylic acid component in order to achieve complete conversion of 2,5-furandicarboxylic acid.

[0117] In another preferred manner, the esterification reaction between 2,5-furandicarboxylic acid and the at least one alcohol is carried out in two stages, wherein already in the first stage substantial amounts of the desired 2,5-furandicarboxylic acid ester are formed without the addition of a catalyst, preferably at least one of the catalysts as described above. The temperatures to be employed in this first stage depend largely on the starting materials. Satisfactory reaction rates are achieved above 100°C, and preferably above 120°C. It is possible to already complete the carboxylic ester formation at these temperatures.

[0118] However, it is more advantageous to increase the temperature continuously up to 160 °C. When using 2,5-furandicarboxylic acid (rather than the corresponding carboxylic anhydride thereof) as the esterification component, the water formed is removed from the reaction system as an azeotrope with the alcohol, as long as the reaction temperature is above the boiling point of the azeotrope (i.e. in a range from 90°C to 100°C under atmospheric pressure). The course and completion of the esterification can in this case be observed via the formation of water. The use of subatmospheric or superatmospheric pressure is not ruled out, but is rather restricted to special cases. To suppress the occurrence of concentration differences, it is advisable to stir the reactor contents or to mix them from time to time, e.g. by passing an inert gas through the reaction mixture.

[0119] It is further optional to work up the formed 2,5-furandicarboxylic acid ester by filtration, optionally followed by distillation.

[0120] In the second stage, the esterification of the 2,5-furandicarboxylic acid is completed. The second stage is carried out in the presence of catalysts at temperatures which are above those employed in the first stage and go up to 250°C. Water formed during the reaction is removed as an azeotrope, with the alcohol acting as an entrainer.

[0121] After completion of the reaction, the reaction mixture comprises not only the desired reaction product, but it may still contain 2,5-furandicarboxylic acid together with excess alcohol and the catalyst.

[0122] To work up the crude 2,5-furandicarboxylic acid ester, the product from the reactor is first neutralized with alkali metal hydroxide or alkaline earth metal hydroxide. The alkaline reagent is employed as an aqueous solution containing from 5 to 20 percent by weight, preferably from 10 to 15 percent by weight, of the hydroxide, based on the overall weight of the solution.

[0123] The amount of neutralizing agent to be used depends on the proportion of acid components, free acid and 2,5-furandicarboxylic acid ester in the crude product. The use of the selected hydroxides, among which sodium hydroxide has been found to be particularly useful, as aqueous solution having a particular concentration and in a defined excess ensures that the acidic constituents of the reaction mixture are precipitated in a crystalline, very readily filterable form.

[0124] At the same time, the catalyst is largely decomposed to form likewise easily filterable products. The alkaline treatment of the crude 2,5-furandicarboxylic acid ester is not tied to the maintenance of particular temperatures. It is advantageously carried out immediately after the esterification step without prior cooling of the reaction mixture.

[0125] Subsequently any free alcohol is separated from the reaction mixture. Steam distillation has been found to be useful for this step and can be carried out in simple form by passing steam into the crude product.

[0126] The removal of the free alcohol is typically followed by the drying of the 2,5-furandicarboxylic acid ester. In a particularly simple and effective embodiment of this step, drying is achieved by passing an inert gas through the product. The crude 2,5-furandicarboxylic acid ester is then filtered to free it of solids. The filtration is carried out in conventional filtration equipment at room temperature or at temperatures up to 150°C. The filtration can also be facilitated by customary filter aids such as cellulose or silica gel.

[0127] Alternatively to the esterification process described above for preparing the 2,5-furandicarboxylic acid esters to be used as lubricants in the present invention, transesterification can also be applied.

[0128] The conventional processes known to the person skilled in the art can be used for the transesterification, e.g. a process that involves the reaction of a di-ester of 2,5-furandicarboxylate, preferably as prepared by the esterification process described above, with at least one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol of the general formula R-OH in the presence of at least one suitable transesterification catalyst, like preferably a titanium(IV) alcoholate.

[0129] Transesterification catalysts that can be used are the conventional catalysts usually used for transesterification reactions, where these are mostly also used in esterification reactions. Among these are by way of example mineral acids, such as sulfuric acid and phosphoric acid; organic sulfonic acids, such as methanesulfonic acid and p-toluenesulfonic acid; and specific metal catalysts from the group of the tin(IV) catalysts, for example dialkyltin dicarboxylates, such as dibutyltin diacetate, trialkyltin alkoxides, monoalkyltin compounds, such as monobutyltin dioxide, tin salts, such as tin acetate, or tin oxides; from the group of the titanium catalysts: monomeric and polymeric titanates and titanium chelates, for example tetraethyl orthotitanate, tetrapropyl orthotitanate, tetrabutyl orthotitanate, triethanolamine titanate; from the group of the zirconium catalysts: zirconates and zirconium chelates, for example tetrapropyl zirconate, tetrabutyl zirconate, triethanolamine zirconate; and also lithium catalysts, such as lithium salts, lithium alkoxides; and aluminum(III) acetylacetonate, chromium(III) acetylacetonate, iron(III) acetylacetonate, cobalt(II) acetylacetonate, nickel(II) acetylacetonate, and zinc(II) acetylacetonate.

[0130] The amount of transesterification catalyst used is from 0.001 to 10% by weight, preferably from 0.05 to 5% by weight. The reaction mixture is preferably heated to the boiling point of the reaction mixture, the reaction temperature therefore being from 20°C to 200°C, depending on the reactants.

[0131] The transesterification can take place at ambient pressure or at reduced or elevated pressure. It is preferable that the transesterification is carried out at a pressure of from 0.001 to 200 bar, particularly from 0.01 to 5 bar. The relatively low-boiling-point alcohol eliminated during the transesterification is preferably continuously removed by distillation in order to shift the equilibrium of the transesterification reaction. The distillation column necessary for this purpose generally has direct connection to the transesterification reactor, and it is preferable that said column is a direct attachment thereto. If a plurality of transesterification reactors is used in series, each of said reactors can have a distillation column, or the vaporized alcohol mixture can preferably be introduced into a distillation column from the final tanks of the transesterification reactor cascade by way of one or more collection lines. The relatively high-boiling-point alcohol reclaimed in said distillation is preferably returned to the transesterification.

[0132] If an amphoteric catalyst is used, this is generally removed via hydrolysis and subsequent removal of the resultant metal oxide, e.g. via filtration. It is preferable that, after reaction has been completed, the catalyst is hydrolyzed by means of washing with water, and the precipitated metal oxide is removed by filtration. The filtrate can, if desired, be subjected to further work-up for the isolation and/or purification of the product. It is preferable that the product is isolated by distillation.

[0133] The transesterification of the di-ester of 2,5-furandicarboxylic acid preferably takes place in the presence of the alcohol component and in the presence of at least one titanium(IV) alcoholate. Preferred titanium(IV) alcoholates are tetrapropoxytitanium, tetrabutoxytitanium, and mixtures thereof. It is preferable that the amount used of the alcohol component is at least twice the stoichiometric amount, based on the di-ester of the 2,5-furandicarboxylic acid used.

[0134] The transesterification can be carried out in the absence of, or in the presence of, an added organic solvent. It is preferable that the transesterification is carried out in the presence of an inert organic solvent. Suitable organic solvents are those mentioned above for the esterification. Among these are specifically toluene and THF.

[0135] The transesterification is preferably carried out in the temperature range from 50 to 200°C.

[0136] The transesterification can take place in the absence of or in the presence of an inert gas. The expression inert gas generally means a gas which under the prevailing reaction conditions does not enter into any reactions with the starting materials, reagents, or solvents participating in the reaction, or with the resultant products. It is preferable that the transesterification takes place without addition of any inert gas.

[0137] One particularly suitable embodiment of the transesterification process comprises:

- a) reaction of 2,5-furandicarboxylic acid with methanol in the presence of concentrated sulfuric acid to give dimethyl 2,5-furandicarboxylate,
- b) reaction of the dimethyl 2,5-furandicarboxylate obtained in step a) with at least one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol of the general formula R-OH in the presence of at least one titanium(IV) alcoholate to give the desired 2,5-furandicarboxylic acid esters to be used as lubricants in the present invention.

[0138] The 2,5-furandicarboxylic acid ester to be used as lubricant in the present invention as well as the lubricant compositions according to the present invention comprising the inventive 2,5-furandicarboxylic acid ester can be favour-

ably used for various applications.

[0139] One preferred application are as components in engine oils. Such general applications includes all sorts of engine oils, including light, medium and heavy duty engine oils, industrial engine oils, marine engine oils, crankshaft oils, compressor oils, refrigerator oils, hydrocarbon compressor oils, very low-temperature lubricating oils and fats, high temperature lubricating oils and fats, wire rope lubricants, textile machine oils, refrigerator oils, aviation and aerospace lubricants, aviation turbine oils, transmission oils, gas turbine oils, spindle oils, spin oils, traction fluids, transmission oils, plastic transmission oils, passenger car transmission oils, truck transmission oils, industrial transmission oils, industrial gear oils, insulating oils, instrument oils, brake fluids, transmission liquids, shock absorber oils, heat distribution medium oils, transformer oils, fats, chain oils, metalworking operations in general, particularly as minimum quantity lubricants for metalworking operations, oil to the warm and cold working, oil for water-based metalworking liquids, oil for neat oil metalworking fluids, oil for semi-synthetic metalworking fluids, oil for synthetic metalworking fluids, drilling detergents for the soil exploration, hydraulic oils, in biodegradable lubricants or lubricating greases or waxes, chain saw oils, release agents, moulding fluids, gun, pistol and rifle lubricants or watch lubricants and food grade approved lubricants.

[0140] In preferred embodiments, the 2,5-furandicarboxylic acid esters of the present invention are used in lubricant compositions, in particular the 2,5-furandicarboxylic acid esters are used in lubricant compositions in automatic transmission fluids, manual transmission fluids, hydraulic fluids, grease, gear fluids, crankcase engine oils, shock absorber fluids, industrial oils, metal-working fluids, transformer oils, biodegradable lubricants and seal plasticizing agents.

[0141] At least 40 mole percent of the 2,5-furandicarboxylic acid, preferably at least 50 mole percent, more preferably at least 65 mole percent, even more preferably at least 75 mole percent, and most preferably at least 85 mole percent of the 2,5-furandicarboxylic acid, and sometimes even at least 90 mole percent, or at least 95 mole percent or even 100 mole percent are hereby derived from a renewable source.

[0142] In a preferred use, the 2,5-furandicarboxylic acid ester is obtained by esterification of the 2,5-furandicarboxylic acid, preferably of the 2,5-furandicarboxylic acid of at least partial renewable origin, with a mixture of Guerbet alcohols as defined herein, which is a mixture that was obtained from the Guerbet reaction. In another preferred embodiment of the present invention, this mixture of Guerbet alcohols is also at least partially derived from a renewable source.

[0143] Accordingly, one very preferred use is directed to the use of the 2,5-furandicarboxylic acid of at least partial, preferably full, renewable origin with a mixture of Guerbet alcohols of at least partially, preferably fully, renewable origin for the preparation of 2,5-furandicarboxylic acid ester of at least partially, preferably fully, renewable origin.

[0144] The 2,5-furandicarboxylic acid esters obtained from esterification of 2,5-furandicarboxylic acid and at least one branched or linear, substituted or unsubstituted aliphatic C6 to C20 alcohol, like a mixture of Guerbet alcohols as described herein, allow the preparation of lubricant compositions with attractive rheological performance characteristics, favourable viscosity profiles, good hydrolytic and oxidative stability, good seal compatibility and favourable traction behaviour.

[0145] Consequently, the lubricant compositions of the present invention due to the presence of the 2,5-furandicarboxylic acid ester have excellent hydrolytic and oxidative stability, good seal performance, and attractive rheological performance characteristics, like kinematic viscosity profile, over a very broad temperature range, and favourable traction behaviour when compared with other lubricants that are based on different carboxylic acid esters. Similarly, the latter performance characteristics of known lubricant compositions can be further improved by the addition, or supplementation of the 2,5-furandicarboxylic acid ester of the present invention.

[0146] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters have kinematic viscosity at 40°C as determined by ASTM D 445 in the range of from 90.0 to 160.0 mm²/s, preferably 100.0 to 150.0 mm²/s, and more preferably 110.0 to 140.0 mm²/s. Most preferred are values in the range of from 120.0 to 135.0 mm²/s.

[0147] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters have kinematic viscosity at 100°C as determined by ASTM D 445 in the range of from 5.0 to 30.0 mm²/s, preferably 10.0 to 25.0 mm²/s, and more preferably 12.0 to 20.0 mm²/s. Most preferred are values in the range of from 15.0 to 18.0 mm²/s.

[0148] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters have viscosity index as determined by ASTM D 2270 in the range of from 125 to 160, preferably 130 to 155, and more preferably from 140 to 150.

[0149] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters have pour point as determined by ISO 3016 in the range of from -70°C to -40°C, preferably from -65°C to -45°C and more preferably from -60°C to -50°C.

[0150] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters have cloud point as determined by ISO 3015 in the range of from -100 to -65, preferably -90 to -70, and more preferably from -85 to -75.

[0151] The lubricant compositions of the present invention comprising the 2,5-furandicarboxylic acid esters also have good oxidation stability and hydrolytic stability.

[0152] The following examples illustrate the invention without being intended to limit the invention thereto.

Examples

General procedure for the synthesis of diester

[0153] To a 2-L round bottom flask fitted with a Dean-Stark apparatus were added 6.00 mol (4.0 eq.) of the respective alcohol (*n*-octanol, 2-ethyl hexanol or 2-propyl heptanol) and 500 g toluene. 2-Propylheptanol is commercially available from BASF SE, Ludwigshafen and represents a mixture of 93.0 wt.-% 2-propyl-heptanol, 2.9 wt.-% 2-propyl-4-methyl-hexanol, 3.9 wt.-% 2-propyl-5-methylhexanol and 0.2 wt.-% 2-isopropylheptanol. The stirred mixture was heated to reflux (114-155 °C) and 234 g (1.50 mol, 1.0 eq.) 2,5-furandicarboxylic acid were added followed by 11.5 g (0.12 mol, 8 mol-%) 99.9% sulfuric acid in 3 or 4 equal portions whenever conversion slowed. The conversion was monitored by the amount of water deposited in the Dean-Stark apparatus. Upon complete conversion a sample was drawn and submitted to GC analysis. The cooled reaction mixture was transferred to a separatory funnel and washed twice with 500 mL saturated NaHCO₃ solution (upon addition of the alkaline NaHCO₃ solution vigorous CO₂ formation may result). The organic phase was washed with brine, dried over anhydrous NaSO₄ and the solvents were removed under reduced pressure. The crude products were purified by fractioning or vacuum distillation or precipitation. The purity and identity of the compounds was determined by GC(-MS) and/or NMR analysis. GC-columns: Agilent J&W DB-5, 30 m x 0,32 mm x 1,0 μm or Ohio Valley OV-1701 60 m x 0,32 mm x 0,25 μm).

[0154] 2-Ethylhexyl-2,5-furandicarboxylate was obtained in 76 % yield and 95.7 % purity 2-Propylheptyl-2,5-furandicarboxylate was obtained in 58 % yield and 97.8 % purity. 2-Octyl-2,5-furandicarboxylate was obtained in 75 % yield and 98.7 % purity.

[0155] Characterization of the diester 2-propylheptyl-2,5-furandicarboxylate

	Units	Methods	Result	
Kinematic viscosity at 40°C	mm ² /s	ASTM D 445	47.7	
Kinetic viscosity at 100°C	mm ² /s	ASTM D 445	5.9	
Viscosity index		ASTM D 2270	46	
Pour point	°C	ISO 3016	-48	
Cloud point	°C	ISO 3015	<-70	
Noack volatility	%	ASTM D5800B	9.3	
Hydrolytic stability	mg KOH/g	SS155181	0 h	0.76
Total acid number			120 h	1.45
			196 h	2.02

[0156] It was found that 2-propylheptyl-2,5-furandicarboxylate was highly viscous while at the same time showing a low pour point. Moreover, 2-propylheptyl-2,5-furandicarboxylate is both hydrolytically and thermally stable.

[0157] The following inventive lubricant composition comprising the 2,5-furandicarboxylic acid ester was prepared and characterized:

Lubricant composition 1	wt.-%
Diester of 2-propylheptyl 2,5-furandicarboxylate (>98,5%)	10.0
Base oil (Synfluid, PAO-6)	52.0
Viscosity modifier, PIB (Lubrizol® 8406, from Lubrizol)	13.0
Viscosity modifier, OCP (Lubrizol® 8407, from Lubrizol)	13.0
Additive package (Anglamol 6004, available from Lubrizol)	12.0

Characterization of lubricant composition 1		
Kinematic viscosity at 40°C (mm ² /s)	ASTM D 445	130,83
Kinematic viscosity at 100°C (mm ² /s)	ASTM D 445	17,668

wherein p is 0, 1, 2, 3 or 4.

5. The use according to claim 3 or 4, **characterized in that** p is 0, 1 or 2.

5 6. The use according to any one of claims 1 to 5, **characterized in that** the 2,5-furandicarboxylic acid and/or the at least one alcohol of the general formula R-OH is at least partially derived from a renewable source.

7. The use according to any one of claims 1 to 6, **characterized in that** at least 40 mole percent of the 2,5-furandicarboxylic acid based on the total amount of 2,5-furandicarboxylic acid in the mixture are derived from a renewable source and/or at least 40 mole percent of the at least one alcohol of the general formula R-OH based on the total amount of the at least one alcohol of the general formula R-OH in the mixture are derived from a renewable source.

8. A lubricant composition comprising the 2,5-furandicarboxylic acid ester according to claims 1 to 7.

15 9. The lubricant composition according to claim 8, further including a base oil component and an additive.

10. The lubricant composition according to claim 9, comprising the following components:

- 5.0 to 25.0 wt.-% of the 2,5-furandicarboxylic acid ester according to claims 1 to 7,
- 40.0 to 90.0 wt.-% of a base oil component,
- 0.1 to 20.0 wt.-% additives,

based on the total weight of the lubricant composition.

25 11. A process for preparing the 2,5-furandicarboxylic acid ester according to any one of claims 1 to 7, comprising the steps of

- providing 2,5-furandicarboxylic acid from a non-renewable source and/or a renewable source,
- preparing a mixture of the 2,5-furandicarboxylic acid and at least one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol of the general formula R-OH derived from a non-renewable source and/or a renewable source,
- carrying out esterification of the 2,5-furandicarboxylic acid and the at least one branched or linear, substituted or un-substituted aliphatic C6 to C20 alcohol of the general formula R-OH.

35 12. Use of the lubricant composition according to any one of claims 8 to 10, in an automatic transmission fluid, a manual transmission fluid, an hydraulic fluid, a grease, a gear fluid, a metal-working fluid, a crankcase engine oil or shock absorber fluid.

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EUROPEAN SEARCH REPORT

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