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(54) Title: POLYLALKENYL SUCCINIMIDES AND USE THEREOF AS DISPERSANTS IN LUBRICATING OILS

(57) Abstract: Polyalkenyl succinimides which can be obtained with a process which comprises: a. reacting a reactive medium-high molecular weight polyalkene with maleic anhydride at a temperature higher than 180°C; b. carrying out the reaction thermally for a time sufficient for having a conversion of the terminal vinylidene groups higher than 10%; c. completing the thermal reaction in the presence of a reaction accelerator consisting of a Lewis acid; d. reacting the reaction product of step (c) with an amine which has at least one primary aminic group capable of forming an imide group.



WO 2009/010226 A1

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POLYALKENYL SUCCINIMIDES AND USE THEREOF AS DISPERSANTS IN
LUBRICATING OILS

The present invention relates to polyalkenyl succinimides and the use thereof as dispersants in lubricating
10 oils.

More specifically, the present invention relates to medium-high-molecular weight polyalkenyl succinimides and the use thereof as dispersants in lubricating oils for internal combustion engines.

15 In the present description, all the conditions indicated in the text should be considered as preferred conditions, even if not expressly declared.

It is known that lubricating oils for internal combustion engines contain numerous additives for reducing or
20 controlling deposits, reducing friction, wear, etc.. The function of dispersants in lubricating oils is to maintain insoluble materials, such as, for example, carbon deposits (soot) and sludge formed by oxidation or due to other complex mechanisms, dispersed in oil. This prevents their
25 flocculation and consequently their precipitation, which

would cause serious damage to the engine.

Furthermore, in diesel engines, dispersants can efficiently contrast the increase in viscosity or the gelification of the lubricant caused by the formation of complex structures which are formed by soot particles which, in the absence of additives, give rise to strong mutual interactions with consequent agglomeration.

Among the known detergent additives, some of the most effective are polyalkenyl acylating agents possibly containing an imide group. These compounds are generally prepared either by classical synthesis, with the use of chlorinated intermediates, or by direct thermal reaction of a polyalkene, characterized by double terminal bonds of the vinylidene type, with an enophyl, at a temperature higher than 150°C, generally higher than 200°C, possibly followed by the reaction with an amine having at least one primary aminic group which can react to form the imide group. Examples of processes for the preparation of these polyalkenyl acylating agents are provided in US patents 3,172,892, 3,912,764, 4,086,251 and 4,152,499.

In the case of products obtained via a thermal process, however, if a chlorine-free product is desired, the preparation processes of the known art are characterized, when carried out at relatively low temperatures (around 200°C), by the drawback of having long reaction times and

rather low yields and, in any case, they provide a product with a low functionalization degree.

The Applicant has now found a dispersant additive for lubricating oils selected from the group of polyalkenyl succinimides, which have a high functionalization degree which makes it particularly effective for this function, as illustrated in the enclosed examples.

An object of the present invention, therefore, relates to polyalkenyl succinimides produced starting from polyalkenyl succinanhydrides having a number average molecular weight, M_n , ranging from about 1,400 to 6,300 and a functionalization degree (FD) higher than 1, generally ranging from 1.30 to 2.50. The functionalization degree is calculated according to the procedure described in US patent 4,952,328.

Alternatively, the functionalization degree of the polyalkenyl succinanhydrides can be calculated more directly and specifically, via H^1 -NMR(FD_{NMR}), by determining, in the reaction mixture, the formation of the various anhydride adducts, using the following formula:

$$FD_{NMR} : (A/2 + B + 2C + 2D) / (A/2 + B + C + D)$$

wherein A and B are the areas of mono-functionalized anhydrides and C and D are those of bis-functionalized anhydrides, calculated from the resonances attributed in the NMR spectra, according to what reported in the literature

(Tessier M., Marechal E.; J. Polym. Sci., Part A: Polym. Chem. 1998; 26: 785-810). In this case the FD_{NMR} of the products can vary from 1 to 2, preferably from 1.3 to 1.9.

More specifically, an object of the present invention
5 relates to polyalkenyl succinimides which can be obtained with a process which comprises:

- a. reacting a reactive polyalkene having a number average molecular weight, M_n , ranging from 1,300 to 6,000 and having a terminal vinylidene groups content higher than or
10 equal to 30%, with an enophyl selected from maleic anhydride or maleic acid at a temperature higher than 180°C;
- b. carrying out the reaction thermally for a time sufficient for having a conversion of the terminal vinylidene groups higher than 10%;
- 15 c. completing the reaction in the presence of a reaction accelerator consisting of a Lewis acid selected from a tin, zinc, aluminum or titanium halides having the formula MX_y , wherein M is the metal, X represents a halide such as chlorine, bromine or iodine, and y varies from 2 to 4. The metallic halide MX_y can also comprise a number of crystalli-
20 zation water molecules ranging from 1 to 5;
- d. reacting the reaction product of step (c), the polyalkenyl succinanehydride, with an amine which has at least one primary aminic group capable of forming an imide group.

25 According to the present invention, the reagent mix-

ture of the first reaction phase (a)-(b) is introduced into a reactor, suitable for performing batch reactions, in any convenient way before heating to the reaction temperature. The reagents, for example, can be charged contemporaneously
5 or sequentially in any order or pre-mixed in a mixing container and then transferred to the reaction vessel. Alternatively, the reaction of phase (a)-(b) can be carried out in continuous.

The reaction is then completed (phase c) in the presence of a reaction accelerator consisting of the Lewis acid
10 selected from a tin, zinc, aluminum or titanium halide. The reaction accelerator is preferably tin chloride $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$.

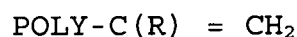
The reaction accelerator is added to the reaction mixture when the conversion of the terminal vinylidene groups
15 of the reactive polyalkene ranges from 15 to 90%, preferably from 30 to 80%. The accelerator is added to the reaction mixture in quantities corresponding to a molar percentage concentration, referred to the reactive double bonds of the polyalkene, ranging from 0.2 to 1.5%, preferably
20 bly from 0.5 to 1.1%.

The reaction of phases (a)-(b) and (c) can be carried out in a solvent and in this case the solvent is a mineral oil able to solubilise reactants and reaction products which can be recovered after bringing the reaction mixture,
25 at the end of the reaction, to room temperature, and after

subjecting it, or not, to filtration. The products recovered are subsequently used for the step (d) of the reaction. Alternatively, step (d) can be performed in the same reaction environment as phases (a)-(c).

5 Phases (a)-(c) can also be carried out in the absence of solvent, subsequently carrying out phase (d) as described above.

The reactive polyalkenes differ from the conventional polyalkenes in the content of terminal vinylidene groups. A
10 polyalkylene chain which has a terminal vinylidene group can be represented by the formula:



wherein R is an alkyl group, whose identity depends on the monomeric unit from which the polyalkene is obtained (for
15 example R is a methyl group for polyisobutene) whereas POLY represents the remaining part of the polyalkenyl chain. Reactive polyalkenes are those which have a content of terminal vinylidene groups equal to at least 35%, preferably equal to at least 40%, more preferably ranging from 50 to
20 95%.

The polyalkenes according to the present invention are generally reactive homopolymers of α -olefins or co-polymers of α -olefins, such as, for example, the ethylene- α -olefin copolymer. Preferred α -olefins according to the present in-
25 vention are those, linear or branched, having the general

formula $\text{CH}_2=\text{CHR}'$ wherein R' represents a linear or branched alkyl radical, containing from 1 to 10 carbon atoms.

Preferred polyalkenes according to the present invention are reactive polyisobutene (PIB) and polybutene-1 with
5 a content of terminal vinylidene groups preferably of at least 45%, for example between 55 and 99%, preferably from 55 to 90%. Methods for the preparation of reactive polyisobutene or polybutene-1 are described, for example, in US patents 4,152,499, 4,605,808, 5,674,955 and in the interna-
10 tional patent application WO 01/19873.

Furthermore, the polyalkenes according to the present invention have a number average molecular weight, M_n , measured, for example, by means of GPC (Gel Permeation Chromatography), osmometry, proton NMR or carbon-13 NMR, ranging
15 from 1,300 to 6,000, for example from 1,500 to 4,000, preferably from 2,000 to 3,000.

Particularly preferred polyalkenes are reactive polyisobutene (PIB) and polybutene-1, mentioned above, and having an average molecular weight M_n ranging from 1,900 to
20 2,400.

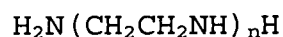
The formation of the intermediate polyalkenyl succin-
anhydride is obtained through the reaction between the re-
active polyalkene and an "enophyl", preferably maleic anhy-
dride or the corresponding acid. Analogous intermediate
25 products, however, obtained from other anhy-

drides/unsaturated polycarboxylic acids, used alone or mixed with the anhydride/maleic acid, fall within the scope of the present invention. Examples of these unsaturated polycarboxylic acids are fumaric acid, itaconic acid, and
5 their corresponding anhydrides or their corresponding C₁-C₄ alkyl esters.

The reaction between the reactive polyalkene and the enophyl takes place at a temperature ranging from 180 to 300°C, preferably from 190 to 250°C and even more preferably
10 bly at about 200°C, at room pressure or under an inert gas, such as nitrogen, at pressures ranging from 0.1 to 1 MPa.

The reactive polyalkylene/enophyl molar ratios generally range from 1:0.9 and 1:3, preferably from 1:1.3 to 1:2.5, for example from 1:1.5 to 1:2.4.

15 The polyalkenyl succinanhydride is reacted with an amine, or a polyamine, having at least one primary aminic group capable of forming an imide group, selected from mono-amines such as methyl amine, 2-ethylhexyl amine, n-dodecyl amine, stearyl amine, etc. or from polyamines such
20 as, for example, propylene diamine. Particularly preferred are polyalkylene polyamines or polyamines having the general formula



wherein n is an integer ranging from 1 to 10. Examples of
25 these polyamines are ethylene diamine, diethylene triamine

(DETA), triethylene tetramine (TETA), tetraethylene pentamine (TEPA), pentaethylene hexamine (PEHA) and higher polyamines (HEPA), etc.

The reaction between the polyalkenyl succinanhydride
5 and the polyamine, for the formation of imides or, in particular, polyisobutenyl succinimides (PIBSI), takes place at a temperature ranging from 100 to 200°C, preferably from 140 to 170°C, at atmospheric pressure, even if it is possible to operate at pressures higher or lower than atmospheric
10 pressure.

The water present in the system or generated during the reaction is preferably removed by stripping under nitrogen. The removal can be facilitated by operating under conditions of reduced pressure.

15 The relative quantities of anhydride and polyamines are selected so that the ratio between the equivalents of succinic anhydride and polyamine (or amine) generally ranges from 0.5 to 2. In this way, by regulating the stoichiometry of the reaction, mono- or disuccinimides can
20 be obtained.

The term imide, as used in the present description and claims, refers to the complete reaction product between the polyamine and the acylating agent (for example polyisobutenyl succinanhydride or PIBSA) which can comprise amides,
25 amidines and possible saline species which can be formed

from the reaction between the anhydride group and amine or polyamine.

The imidation reaction can take place in the same reaction environment as the anhydride, or the polyalkenyl succinanhydride can be recovered with the known methods, 5 for example by gear pumps, and can be transferred and reacted separately with the amine. The reaction can be carried out in the presence or absence of a solvent (mass imidation) and in the latter case the solvent can be added at 10 the end. In this way, the final succinimide can be filtered, if necessary, moved, stored or more advantageously mixed with other components.

Solvents suitable for the purpose are mineral or synthetic oils having a viscosity suitable for being used in 15 internal combustion engines. Mineral oils for use as diluent according to the present invention include paraffinic oils, naphthenic oils and other oils normally used as components of lubricating oils. Synthetic oils can include mixtures of hydrocarbons consisting, for example, of polymers of alpha-olefins having a suitable viscosity, for ex- 20 ample liquid hydrogenated oligomers of C₆-C₁₂ alpha-olefins, such as trimers of 1-decene. Blends of synthetic oils can also be used.

The polyalkenyl succinimides, object of the present 25 invention, can be used as dispersant additives for lubri-

cating oils or they can be used together with other components in "additive concentrates" or "packages" to be added to the lubricating oil whose performances are to be improved. The final dispersant is a highly desirable alternative with respect to analogous dispersants produced according to the classical synthesis process starting from the synthesis of anhydrides by means of chlorinated intermediates.

Furthermore, the polyalkenyl succinimides, object of the present invention, generally have a higher functionalization degree than that obtained with the "classical" thermal reaction, which makes them more reactive with respect to the dispersants obtained with non-catalytic thermal processes.

A further object of the present invention relates to hydrocarbon compositions comprising polyalkenyl succinimides prepared as described above.

In particular, a further object of the present invention relates to hydrocarbon lubricating compositions comprising:

- i. one or more lubricant base oils having a viscosity ranging from 3 to 15 cSt at 100°C, preferably from 5 to 10 cSt at 100°C, suitable for being used as automotive lubricants; and
- ii. from 0.1 to 15% by weight with respect to the total

composition, preferably from 1 to 10%, of a polyalkenyl succinimide, deriving from a polyalkenyl succinanehydride having a number average molecular weight M_n ranging from about 1,400 to 5,300 and a functionalization degree (FD) higher than 1.0 or FD_{NMR} ranging from 1 to 2, obtained with the process comprising steps (a)-(d) described above.

The hydrocarbon composition object of the present invention can also contain conventional additives used in automotive lubricants such as detergents, friction-modifiers, antioxidants, etc.

Particularly preferred in the hydrocarbon compositions of the present invention are polyisobutenyl succinimides, deriving from polyisobutenyl succinanehydrides having a number average molecular weight M_n ranging from 2,000 to 3,000 and a functionalization degree (FD) higher than 1.0 or FD_{NMR} ranging from 1 to 2, obtained with the process comprising steps (a)-(d) described above.

Non-conventional or synthetic mineral oils, belonging to groups I-V according to the API (American Petroleum Institute) classification, used individually or in blends, fall within the definition of lubricating base oils and are preferred.

The mineral oils can consist of mixtures of paraffinic oils, naphthenic oils and other oils normally used as com-

ponents of automotive engine oils.

The synthetic oils can include, for example, hydrocarbon mixtures consisting of polymers of alpha-olefins having a suitable viscosity, for example liquid hydrogenated oligomers of C₆-C₁₂ alpha-olefins, such as trimers of 1-decene, etc.

The lubricating base oils can be used alone or in blends.

The present invention is now illustrated for illustrative and non-limiting purposes by the following examples.

EXAMPLE 1

Synthesis of polyisobutenyl succinanhydride (PIBSA)

In a generic experiment, 100 g of PIB (Glissopal 2,300, BASF; Mn 2,300) are introduced into a 250 mL cylindrical glass reactor, equipped with a mechanical stirrer and a reflux condenser.

The reactor is flushed with nitrogen, under stirring, until a temperature of 110°C is reached. After 30 minutes, 6.39 g of maleic anhydride (MA) are added, under nitrogen, and the mixture is heated to 200°C.

After at least 50% of the reaction has been completed, SnCl₂·2H₂O is added, under stirring, and the reaction is kept under stirring for twenty-one hours. The temperature is then lowered to 160°C and the unreacted enophyl is

stripped under vacuum (0.2 mm Hg).

The weight conversion degree of the reaction was evaluated, by difference, by quantifying the weight of unreacted polyisobutylene after its separation from the reaction mixture. A weighed quantity of polyisobutenyl succin
5 nanhydride (PIBSA), dissolved in n-heptane, is eluted through a chromatographic column, containing silica gel. The eluted phase, containing only unreacted PIB, is then evaporated, dried under vacuum (0.2 mm Hg) and weighed.

10 The functionalization degree (FD), expressed as grafted moles of succinic anhydride per mole of reacted polymer, was determined according to the procedure described in US patent 4,952,328, after determining the acidity of the PIBSA, by titration according to what is de
15 scribed in the method ASTM D 664

The NMR functionalization degree (FD_{NMR}) was calculated after determining the adducts present in the PIBSA according to the method described in literature: Tessier M., Marechal E. J. Polym. Sci., Part A: Polym. Chem. 1988; 26;
20 785-810.

Table 1 shows the yields and the functionalization degree of the products, obtained at various times, in the presence and absence of catalyst.

25

Table 1

PIB Mn	T (°C)	Cat. (mol.%)	PIB/MAH	reac. time (hrs)	Yield (w%)	FD	FD _{NMR}
2300	200	0.00	1/1.5	7	67.5	1.62	1.07
				10	71.3	1.73	1.11
				12	73.0	1.76	1.18
				21	75.8	1.76	1.20
2300	200	0.5 (1)	1/1.5	7	69.7	1.65	1.05
				10	72.8	1.92	1.47
				12	74.3	1.84	1.49
				21	77.1	2.05	1.52

(1) Catalyst added after partial conversion of PIB

Synthesis of polyisobutenyl succinimide

The polyisobutenyl succinimide (PIBSI) is prepared by introducing 100 g of polyisobutenyl succinanhdyride (PIBSA) into a jacketed cylindrical glass reactor, equipped with a mechanical stirrer, a lower drainage valve and a reflux condenser. After the addition of 110 g of base oil SN150, 7.23 g of HEPA are added at a temperature of 130°C, under a nitrogen blanket, the temperature is increased to 165°C and the reaction is continued for about two hours. The water formed is stripped away by applying, for an hour, a nitrogen flow from the bottom of the reactor.

EXAMPLE 2

Evaluation of the products: Spot Test

The evaluation of the dispersing properties of the products obtained in example 1, can be performed by means of the "spot test". In this test, a lubricant formulated with 8% by weight of the dispersant to be analyzed, is
5 mixed, at room temperature, with synthetic sludges and stirred twice, by using a turbine, at about 20,000 rev/min for 30 seconds with a 15 seconds interval.

After heating to 250°C for 10 minutes, the dispersion thus obtained is left to cool to room temperature. An ali-
10 quot of the dispersion, collected by means of a pipette, is deposited on a filter paper sheet, prepared in advance and fixed to a frame, forming a more or less circular spot. The filter paper is then placed, for 24 hours, in a thermostated oven set at 40°C.

15 The evaluation of the dispersing properties of the formulate under examination is carried out, performing duplicate tests, by analyzing and quantifying the spot produced on the filter paper. This is obtained by determining a coefficient, ID%, calculated by normalizing to 100 the
20 ratio between the inner and outer diameters of the spot halos according to the following relationship:

$$ID\% = (ID/ED) \cdot 100$$

wherein ID and ED are respectively the inner and the outer spot diameters. The higher the ratio under examination, the
25 greater the dispersing efficacy of the oil, as the exten-

sion of the inner diameter is related to the dispersion of the sludge.

Table 2 shows the results obtained with a SAE 5W40 lubricating oil formulated with the PIBSI of example 1 at 8% by weight. The results are compared with those obtained with a commercial oil of analogous SAE grade.

The data of the laboratory test, demonstrate the dispersant efficacy, with respect to the sludge, of the additive object of the present invention when compared with the commercial product.

Table 2

		Stain 1			Stain 2			
Sample	SAE	ID (cm)	ED (cm)	ID%	ID (cm)	ED (cm)	ID%	Average ID%
commercial oil	5w40	1.5	4.0	37.5	1.8	4.5	40.0	38.8
oil + PIBSI	5w40	1.9	3.4	56.0	1.8	3.2	56.3	56.1

Results obtained with the CEC L-93-04 engine test.

The CEC L-93-04 engine test (dispersion at medium temperature), was used for evaluating the dispersing properties of an oil after the addition of the dispersant of example 1.

The above mentioned method is part of the ACEA 2004 (Association des Constructeurs Europeen d'Automobiles) specifi-

cations, and simulates the behaviour of passenger car diesel engines under high-speed highway service.

The test is carried out using a Peugeot diesel engine DV4TD, equipped with a "common rail" injection system, which is run continuously for 120 hours within a standard procedure. The performances of the lubricant are evaluated at a 24 hours time interval, determining at different times, the increase in the kinematic viscosity at 100°C with respect to the content of carbonaceous residue (soot).

The values of these parameters are used to calculate, by interpolation, the absolute viscosity increase found in correspondence with a 6% soot content. This value is then compared with the equivalent value found for a reference oil (RL223).

Pass / Fail criteria

A positive result (PASS) is obtained when the viscosity percentage increase of the oil under examination, determined at a 6 % soot concentration does not exceed 60% of the analogous value obtained with the reference oil.

The polyisobutenyl succinimides prepared according to example 1, were introduced in a suitable "package" containing other additives and added to an appropriate base oil in such a way that a SAE 5W40 lubricant, according to the SAE J300 classification, with a final 8% (wt/wt) dispersants concentration was obtained.

Table 3 shows the results obtained in the DV4TD test.

Table 3

Formulate	5w40	A3/B3-04	Result
SAE Grade		Limits	
Absolute viscosity increase at 100°C and 6% of soot (cSt)	4.66	8.74	PASS

The value of the viscosity percentage increase, at 6% of soot, obtained in the motor engine test, shows that the lubricant oil formulated with the dispersant object of the present invention, brilliantly passes the standard engine test, demonstrating the efficacy of the polyalkenyl succinimide used.

CLAIMS

1. Polyalkenyl succinimides deriving from polyalkenyl succinanhydrides with a functionalization degree higher than 1.0 or a FD_{NMR} ranging from 1 to 2 obtained with a
5 process comprising:

a. reacting a reactive polyalkene having a number average molecular weight, M_n , ranging from 1,300 to 6,000 and having a content of terminal vinylidene groups higher than or equal to 30%, with an enophyl selected from anhydride or
10 maleic acid, at a temperature higher than 180°C;

b. carrying out the reaction thermally for a time sufficient for having a conversion of the terminal vinylidene groups higher than 10%;

c. completing the thermal reaction in the presence of a reaction accelerator consisting of a Lewis acid selected from
15 a tin, zinc, aluminium or titanium halides having the formula MX_y , wherein M is the metal, X represents a halide such as chlorine, bromine or iodine, and y varies from 2 to 4;

20 d. reacting the reaction product of step (c), the polyalkenyl succinanhydrides, with an amine which has at least one primary aminic group capable of forming an imide group.

2. The polyalkenyl succinimides according to claim 1, wherein the reactive polyalkenes are reactive homopolymers
25 of α -olefins or α -olefins copolymers.

3. The polyalkenyl succinimides according to claim 2, wherein the reactive polyalkenes are polyisobutene (PIB) or polybutene-1 having a content of terminal vinylidene groups of at least 45% and a number average molecular weight ranging from 1,900 to 2,400.

4. The polyalkenyl succinimides according to any of the previous claims, wherein the reaction between the reactive polyalkene and the enophyl takes place at a temperature ranging from 180 to 300°C, at atmospheric pressure or under a pressure of inert gas ranging from 0.1 to 1 MPa.

5. The polyalkenyl succinimides according to any of the previous claims, wherein the reactive polyalkylene/enophyl molar ratios range from 1:0.9 to 1:3.

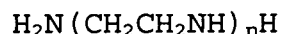
6. The polyalkenyl succinimides according to any of the previous claims, wherein the reaction accelerator is added to the reaction mixture when the conversion of the terminal vinylidene groups of the reactive polyalkene ranges from 20 to 90%.

7. The polyalkenyl succinimides according to any of the previous claims, wherein the accelerator is added to the reaction mixture in quantities corresponding to a molar percentage concentration, referring to the reactive double bonds of the polyalkene, ranging from 0.2 to 1.5%.

8. The polyalkenyl succinimides according to any of the previous claims, wherein the metallic halide MX_y comprises

a number of crystallization water molecules ranging from 1 to 5.

9. The polyalkenyl succinimides according to any of the previous claims, wherein the polyamines are selected from those having the general formula



wherein n is an integer ranging from 1 to 10.

10. The polyalkenyl succinimides according to any of the previous claims, wherein the reaction between the polyalkenyl succinanthydride and the polyamine takes place at a temperature ranging from 100 to 200°C.

11. The polyalkenyl succinimides according to any of the previous claims, wherein the relative quantities of anhydride and polyamines are selected so that the ratio between the equivalents of succinic anhydride and those of the polyamine (or amine) ranges from 0.5 to 2.

12. A hydrocarbon lubricating composition comprising:

- i. one or more lubricating base oil having a viscosity ranging from 3 to 15 cSt at 100°C, suitable for being used in automotive engine oils ; and
- ii. from 0.1 to 15% by weight, with respect to the total composition of a polyalkenyl succinimide, deriving from a polyalkenyl succinanthydride having a number average molecular weight M_n ranging from about 1,400 to 5,300 and a functionalization degree (FD) higher than 1.0 or FD_{NMR} ranging

from 1 to 2, obtained with the process comprising steps (a) - (d) described above.

13. The hydrocarbon lubricating composition according to claim 12, wherein polyalkenyl succinimide is selected from polyisobutenyl succinimides, obtained from polyisobutenyl succinanhydrides having a number average molecular weight, M_n , ranging from about 2,000 to 3,000 and a functionalization degree (FD) higher than 1.0 or FD_{NMR} ranging from 1 to 2, substantially obtainable by means of the process comprising steps (a) - (d) previously described.

14. The hydrocarbon lubricating composition according to claims 12 or 13, wherein the lubricating base oil consists of mineral oils belonging to groups I-V according to the API (American Petroleum Institute) classification, used individually or in a blend.

INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2008/005629

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08F8/46 C08F8/32 C08F10/00 C10M133/56 C10M169/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08F C10M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, COMPENDEX

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 97/47666 A (BP CHEM INT LTD [GB]) 18 December 1997 (1997-12-18) claims 1,3-7,9-12,14-22,27 page 1, lines 4-22 page 4, lines 7-32 page 5, lines 1,2 page 6, lines 11-33 example 8; table 4	1-14
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☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search

31 October 2008

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2008/005629

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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International application No

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