

VISCOSITY INDEX IMPROVER ADDITIVES OF LUBRICATING OILS AND PROCESS FOR THEIR PREPARATION

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DESCRIPTION

5 The present invention relates to viscosity index improver (V.I.I.) additives of lubricating oils.

More particularly, the present invention relates to a viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene, said hydrogenated linear polybutadiene having the specific characteristics reported below.

10 The aforesaid viscosity index improver (V.I.I.) additive of lubricating oils is advantageously usable in compositions of lubricating oil such as, for example, compositions of lubricating oil for both diesel and petrol passenger cars, or for heavy duty diesel engines, in functional fluids such as, for example, fluids for manual or automatic transmission, hydraulic fluids, and in industrial lubricants (for example, gear
15 lubricants for wind turbines).

The present invention also relates to processes for the preparation of said hydrogenated linear polybutadiene.

The present invention also relates to concentrated solutions comprising at least one viscosity index improver (V.I.I.) additive of lubricating oils reported above in at
20 least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, synthetic origin, or mixtures thereof.

The present invention also relates to a composition of lubricating oil comprising at least one viscosity index improver (V.I.I.) additive of lubricating oils reported above, as such or in concentrated solution, and at least one lubricating base oil, said lubricating
25 base oil being selected from lubricating base oils of mineral origin, of synthetic origin, or mixtures thereof.

It is known that the viscosity of the lubricating oils varies with temperature. Many lubricating oils must in fact be used in a wide temperature range and therefore it is important that the oil is not too viscous at low temperature and is not too fluid at high
30 temperature. The viscosity variation of a lubricating oil with temperature is expressed by the value of the viscosity index: the higher the viscosity index value, the lower the change in viscosity of the lubricating oil with temperature.

The use of polymer-based additives as viscosity index improvers (V.I.I.) of the lubricating oils is also known, in order to modulate the viscosity of said lubricating oils as the temperature varies, i.e. to increase the viscosity at high temperature and to limit as much as possible the increase in viscosity at low temperature.

5 For example, polymers usually used as viscosity index improvers (V.I.I.) of the lubricating oils are: ethylene-propylene copolymers [also known in the art as olefin copolymers (OCP)], hydrogenated conjugated polydienes (for example, hydrogenated polyisoprene), hydrogenated styrene/butadiene copolymers, hydrogenated styrene/isoprene copolymers, polyalkylmethacrylates, diene/alkylmethacrylate
10 copolymers.

The synthesis and use in lubricants of hydrogenated linear polymers of conjugated dienes and of styrene-conjugated dienes copolymers in compositions of lubricating oil are described, for example, in the following patents: US 3,554,911 (hydrogenated butadiene-styrene random copolymers); US 3,668,125 (hydrogenated or saturated block
15 copolymers having at least three blocks, for example, polystyrene-hydrogenated polybutadiene-polystyrene, polystyrene-hydrogenated polyisoprene-polystyrene); US 3,772,196 [di-block copolymer comprising a first block deriving from an alkenyl arene (e.g., styrene) and a second block deriving from essentially completely hydrogenated isoprene]; US 3,775,329 ("tapered" hydrogenated isoprene-styrene copolymer); US
20 3,835,053 (hydrogenated polysoprene homopolymer); EP 585269 [hydrogenated butadiene-diene copolymers, e.g. hydrogenated butadiene-isoprene copolymer, possibly with a star structure] and EP 578725 (hydrogenated polybutadiene block copolymer comprising 1,4-butadiene monomer units and 1,2-butadiene monomer units).

The synthesis of ethylene-propylene copolymers, as well as lubricating
25 compositions containing them are described, for example, in patents EP 3,950,897, EP 3,950,892, EP 3,950,899, EP 3,950,898, EP 3,950,902, EP 3,950,900, EP 3,950,895, EP 3,950,893, EP 3,950,901 and EP 3,950,894.

Furthermore, since low molecular weights are indispensable to guarantee the application performance of the viscosity index improvers (V.I.I.) of the lubricating oils
30 and the processes usually used for the production of copolymers and terpolymers of ethylene [EP(D)M)] (solution or slurry processes) often provide copolymers and terpolymers of ethylene [EP(D)M)] having significantly higher molecular weights, it is

necessary to subject said copolymers and terpolymers of ethylene [EP(D)M)] to thermodegradation processes as described, for example, in patents EP 1,013,673 and US 6,753,381.

The synthesis of hydrogenated styrene-diene copolymers with a stellar structure
5 *in situ* in low viscosity base oil, as well as compositions of lubricating oil containing them are described, for example, in patent CN 113461881.

The synthesis of diene-alkyl methacrylate copolymers, in conventional solvents (for example, toluene, 1-decene, cyclohexane) is described, for example, in patents EP 3,880,774 and EP 3,498,808.

10 A composition of lubricating oil comprising low molecular weight polymethacrylates is described, for example, in EP 3,510,131.

For each of the aforesaid classes of (co)polymers, the increase in the weight average molecular weight (M_w) corresponds to an increase in the thickening power (TP) and therefore the amount of (co)polymer necessary to obtain a certain increase in the
15 viscosity index of the high temperature composition of lubricating oil (thickening) is reduced. A (co)polymer, in order to be a good viscosity index improver (V.I.I.) additive, must not only have a beneficial influence on the viscosity index of the fresh lubricating oil, but it must also be stable and maintain its function even when the lubricating oil is in use in an engine and, for this reason, a good viscosity index improver (V.I.I.) additive
20 must also have good mechanical shear stability [i.e. good values of the shear stability index (SSI)]. It is known that, contrary to the thickening power (TP), the mechanical shear stability of a (co)polymer decreases with the increase of its weight average molecular weight (M_w) and therefore the selection of a viscosity index improver (V.I.I.) additive is usually a compromise between the use of high amounts of (co)polymers with
25 low weight average molecular weight (M_w) that are stable to mechanical shear [i.e. having low value of the shear stability index (SSI)] but having low thickening power (TP) and the use of small amounts of (co)polymers with high weight average molecular weight (M_w) that are poorly stable to mechanical shear [i.e. having high value of the shear stability index (SSI)] but having high thickening power (TP).

30 However, the above reported processes can have some drawbacks. For example, in order to obtain ethylene-propylene copolymers having molecular weights suitable for use as viscosity index improvers (V.I.I.), it can be necessary to use hydroperoxides, or

operate at high temperatures (up to 500°C) that require operation in an inert atmosphere (for example, in the presence of nitrogen) and that can lead to the formation of gels and branchings in the final product. Furthermore, in order to improve the dimensional stability of the ethylene-propylene copolymers obtained, it can be necessary to use small
5 amounts of polyvinylarene/hydrogenated conjugated diene/polyvinylarene block copolymers or polyfunctional vinyl monomers.

In addition, the ethylene-propylene copolymers obtained from Ziegler-Natta polymerization often have a polydispersity index greater than 2 and this implies, in order to maximize mechanical shear stability, the need to synthesize ethylene-propylene
10 copolymers having an extremely low weight average molecular weight (M_w) at the expense of the viscosity index.

The Applicant has therefore posed the problem of finding viscosity index improver (V.I.I.) additives of lubricating oils capable of presenting excellent values of shear stability index (SSI) and good fluidity at low temperature (“Pour Point”).

15 The Applicant has now found a viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene having the specific characteristics reported below. The aforesaid viscosity index improver (V.I.I.) additive of lubricating oils is advantageously usable in compositions of lubricating oil such as, for example, compositions of lubricating oil for both diesel and petrol passenger cars, or
20 for heavy duty diesel engines, in functional fluids such as, for example, fluids for manual or automatic transmission, hydraulic fluids, and in industrial lubricants (for example, gear lubricants for wind turbines). The aforesaid viscosity index improver (V.I.I.) additive of lubricating oils is therefore particularly useful in the engine sector where correct lubrication is increasingly important both to prolong the life of the engine,
25 especially the diesel one, but also in the latest direct injection petrol engines, and to reduce fuel consumption: said correct lubrication therefore has a direct benefit on the sustainability and reduction of engine emissions. In addition, the aforesaid viscosity index improver (V.I.I.) additive of lubricating oils is particularly useful in sectors where excellent mechanical shear stability is required [(i.e. good shear stability index (SSI) values] and good low-temperature properties [i.e. good low-temperature fluidity (“Pour Point”)].
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The object of the present invention is therefore a viscosity index improver (V.I.I.)

additive of lubricating oils comprising a hydrogenated linear polybutadiene, said hydrogenated linear polybutadiene having the following characteristics:

- weight average molecular weight (M_w) comprised between 1000 Dalton and 40000 Dalton, preferably comprised between 1500 Dalton and 38000 Dalton;
- 5 - a content of units having configuration 1,2 comprised between 40% by weight and 80% by weight, preferably comprised between 45% by weight and 75% by weight, with respect to the total amount of butadiene units present in said hydrogenated linear polybutadiene;
- a polydispersion index (PDI), that is, the ratio between the weight average molecular weight (M_w) and the number average molecular weight (M_n) (M_w/M_n),
10 less than or equal to 1.3, preferably comprised between 1 and 1.1;
- a degree of hydrogenation greater than or equal to 98%, preferably comprised between 98.5% and 100%;
- a dynamic viscosity, measured at 40°C, comprised between 5 Pa*s and 10000
15 Pa*s, preferably comprised between 10 Pa*s and 8000 Pa*s;
- a dynamic viscosity, measured at 100°C, comprised between 0.1 Pa*s and 1000 Pa*s, preferably comprised between 0.2 Pa*s and 500 Pa*s;
- an enthalpy of crystallization (ΔH_c), determined by temperature modulated differential scanning calorimetry (TMDSC), comprised between 0 J/g and 1 J/g,
20 preferably comprised between 0 J/g and 0.8 J/g.

For the purpose of the present description and the following claims, the definitions of the numerical ranges always include the extreme values unless otherwise specified.

For the purpose of the present description and the following claims, the term “comprising” also includes the terms “which essentially consists of” or “which consists
25 of”.

For the purpose of the present description and the following claims, the content of units having 1,2 configuration of the polybutadiene, said content being expressed as % by weight with respect to the total amount of the butadiene units present in the hydrogenated linear polybutadiene, was determined as reported below in the paragraph
30 “Spectra $^1\text{H-NMR}$ ” on the non-hydrogenated polybutadiene (“parent polymer”).

In accordance with a preferred embodiment of the present invention, said viscosity index improver (V.I.I.) additive of lubricating oils has the following characteristics:

- a shear stability index (SSI) comprised between 0% and 7%, preferably comprised between 0% and 5%, measured in accordance with the CEC-L-14-A-93 standard (30 cycles of shearing) in Group III 4 cSt base oil;
- a pour point lower than -30 C, preferably comprised between -33 C and -45°C, measured in accordance with the ASTM D5950-14 standard in Group III 4 cSt base oil in the presence of the additive Viscoplex[®] V1-254 (Evonik) (Pour Point Depressant - PPD).

In accordance with a preferred embodiment of the present invention, said viscosity index improver (V.I.I.) of lubricating oils in order to reach a target dynamic viscosity measured in accordance with the ASTM D7042-04 standard, at 100 C, in Group III 4 cSt base oil comprised between 6 cSt and 9 cSt, preferably comprised between 7 cSt and 8 cSt, is used in said Group III 4 cSt base oil at a target concentration comprised between 1.9% by weight and 12% by weight, preferably comprised between 2.1% by weight and 10% by weight, with respect to the total weight of the Group III 4 cSt base oil and viscosity index improver (V.I.I.) of lubricating oils composition.

As mentioned above, the present invention also relates to processes for the preparation of the aforesaid hydrogenated linear polybutadiene.

It is therefore a further object of the present invention, a first discontinuous (batch) process for the preparation of a hydrogenated linear polybutadiene comprising the following steps:

- (a) subjecting 1,3-butadiene to living polymerization by an anionic route in the presence of at least one hydrocarbon solvent, at least one lithium-based initiator and at least one polar modifier and continuing said polymerization until substantially complete conversion of the 1,3-butadiene;
 - (b) adding at least one termination agent to the polymerization mixture obtained in step (a);
 - (c) subjecting the linear polybutadiene obtained in step (b) to hydrogenation obtaining a mixture comprising hydrogenated linear polybutadiene;
 - (d) subjecting the mixture comprising hydrogenated linear polybutadiene obtained in step (c) to desolventisation and recovering the hydrogenated linear polybutadiene;
- wherein said desolventisation step (d) is carried out in the absence of water.

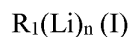
For the purpose of the present description and the following claims, the phrase

“substantially complete conversion” means that the polymerization has continued until at least 98%, preferably at least 99%, more preferably 100%, of the loaded monomer, i.e. 1,3-butadiene, was polymerized.

In accordance with a preferred embodiment of the present invention, said at least one hydrocarbon solvent can be selected, for example, from aliphatic, cycloaliphatic or aromatic hydrocarbon solvents such as, for example, propane, *n*-butane, *iso*-butane, *n*-pentane, *iso*-pentane, *n*-hexane, *n*-heptane, *n*-octane, cyclohexane, cyclopentane, propene, 1-butene, *iso*-butene, *trans*-2-butene, *cis*-2-butene, 1-pentene, 2-pentene, 1-hexene, 2-hexene, benzene, toluene, xylene, ethylbenzene, or mixtures thereof. Said solvents can be advantageously used in anhydrous form. Cyclohexane, *n*-hexane, or mixtures thereof, in anhydrous form, are preferred.

Generally, the amount of hydrocarbon solvent used in said copolymerization by an anionic route is such as to allow complete solubility of the monomer (i.e. 1,3-butadiene), of the additives optionally present, of the compounds obtained in the aforesaid steps (a)-(d), the complete stirring of the reaction mixture, also during said copolymerization, and the diffusion of the heat of reaction. Preferably, said hydrocarbon solvent is used in such an amount as to have a monomer concentration (i.e. 1,3-butadiene) in the hydrocarbon solvent comprised between 4% by weight and 20% by weight, more preferably between 6% by weight and 15% by weight, with respect to the total weight of the hydrocarbon solvent.

In accordance with a preferred embodiment of the present invention, said at least one lithium-based initiator can be selected, for example, from compounds having general formula (I):



wherein R_1 represents an alkyl group C_1 - C_{20} , preferably C_2 - C_{12} , linear or branched, a cycloalkyl group C_3 - C_{30} , preferably C_4 - C_{10} , an aryl group C_6 - C_{30} , preferably C_6 - C_{12} , and n is an integer comprised between 1 and 4.

In accordance with a particularly preferred embodiment of the present invention, said at least one lithium-based initiator can be selected, for example, from: lithium methyl, lithium *n*-butyl, lithium *sec*-butyl, lithium *t*-butyl, lithium *n*-propyl, lithium *iso*-butyl, lithium amyl, lithium cyclohexyl, lithium phenyl, lithium 1-methyl styryl, lithium *p*-tolyl, lithium naphthyl, L,L-diphenyl-5-lithium-3-methyl-pentyl, or mixtures thereof.

Lithium *n*-butyl is preferred.

The amount of lithium-based initiator that can be used in the process object of the present invention depends on various factors such as, for example, the molecular weight of the polymer to be obtained, the impurities optionally present in the polymerization
5 mixture. Generally, said lithium-based initiator can be used in an amount comprised between 0.16 phm and 6.4 phm, preferably in an amount comprised between 0.17 phm and 4.3 phm, (phm = parts per hundred parts by weight of monomer).

In accordance with a preferred embodiment of the present invention, said at least one polar modifier can be selected, for example, from: non-cyclic ethers such as, for
10 example, ethyl ether, or mixtures thereof; tertiary amines such as, for example, tri-butyl amine; cyclic ethers such as, for example, tetrahydrofuran (THF); chelating ethers such as, for example, ethylene glycol dimethyl ether (dimethylglyme), dioxane, 2-methoxy-ethyl-tetrahydrofuran (THFA-ethyl), 2-methoxy ethyl-tetrahydropyran, or mixtures thereof; chelating amines such as, for example, *N,N,N',N'*-
15 tetramethylenethylenediamine (TMEDA); or mixtures thereof. Tetrahydrofuran (THF), 2-methoxy-ethyl-tetrahydrofuran (THFA-ethyl), or mixtures thereof, are preferred.

The amount of polar modifier that can be used in said first process object of the present invention depends on various factors such as, for example, the type of polar modifier used and the type of lithium-based initiator used. By way of example, in the
20 case of the aforesaid first process object of the present invention, tetrahydrofuran (THF) can be used in an amount comprised between 10000 ppm and 35000 ppm, preferably comprised between 11000 ppm and 30000 ppm (ppm = parts per million), with respect to the hydrocarbon solvent used.

In accordance with a preferred embodiment of the present invention, said at least
25 one termination agent can be selected, for example, from trimethyl chlorosilane, butanol, octanol, or mixtures thereof. Trimethyl chlorosilane is preferred. Preferably, said termination agent and said lithium-based initiator can be used in a molar ratio comprised between 0.8 and 1.3, more preferably comprised between 0.9 and 1.2.

In accordance with a preferred embodiment of the present invention, in said first
30 process:

- said step (a) can be carried out at a temperature comprised between 20°C and 100°C, preferably comprised between 25°C and 85°C; and/or for a time comprised

between 10 minutes and 1 hour, preferably comprised between 15 minutes and 40 minutes; and/or

- said step (b) can be carried out at a temperature comprised between 30°C and 115°C, preferably comprised between 40°C and 110°C, and/or for a time
5 comprised between 1 minute and 30 minutes, preferably comprised between 3 minutes and 20 minutes.

The aforesaid hydrogenation step (c) can be carried out in accordance with processes known in the art. Preferably, it can be carried out in the presence of a titanium/magnesium-based catalyst composition (for exemplar, bis-cyclopentadienyl-
10 titanium-dichloride/butyl-ethyl-magnesium) as described, for example, in European patents EP 0816382 and EP 0914867.

The aforesaid desolventisation step (d) in the absence of water can be carried out, for example, by vacuum evaporation, or by means of a stirred thin film evaporator.

The hydrogenated linear polybutadiene obtained at the end of the aforesaid process
15 can be stored as such or solubilized in an oil of mineral or synthetic origin before being stored.

A further object of the present invention is a second discontinuous (batch) process, by an anionic route, for the preparation of a hydrogenated linear polybutadiene comprising the following steps:

- 20 (a') subjecting 1,3-butadiene to living polymerization by an anionic route in the presence of at least one lubricating base oil, at least one lithium-based initiator and at least one polar modifier and continuing said polymerization until substantially complete conversion of the 1,3-butadiene;
- (b') adding at least one termination agent to the polymerization mixture obtained in
25 step (a');
- (c') subjecting the linear polybutadiene obtained in step (b') to hydrogenation, obtaining a mixture comprising hydrogenated linear polybutadiene.

The mixture obtained at the end of step (c') can comprise hydrogenated linear polybutadiene in an amount comprised between 1% by weight and 70% by weight,
30 preferably comprised between 1.5% by weight and 50% by weight, with respect to the total weight of the mixture. Said mixture can be stored as such.

In accordance with a preferred embodiment of the present invention, in said

second process:

- said step (a') can be carried out at a temperature comprised between 20°C and 100°C, preferably comprised between 25°C and 85°C; and/or for a time comprised between 10 minutes and 1 hour, preferably comprised between 20 minutes and 40 minutes; and/or
- said step (b') can be carried out at a temperature comprised between 30°C and 115°C, preferably comprised between 40°C and 110°C, and/or for a time comprised between 1 minute and 30 minutes, preferably comprised between 3 minutes and 20 minutes.

10 The aforesaid hydrogenation step (c') can be carried out as described above in accordance with processes known in the art. Preferably, it can be carried out in the presence of a titanium/magnesium-based catalyst composition (for example, bis-cyclopentadienyl-titanium-dichloride/butyl-ethyl-magnesium) as described, for example, in European patents EP 0816382 and EP 0914867.

15 It should be noted that in accordance with said second process the desolventisation step is eliminated with a consequent saving of time and costs.

Said at least one lithium-based initiator, at least one polar modifier, and at least one termination agent can be selected from those reported above.

20 Said at least one lubricating base oil can be selected from the lubricating base oils of mineral or synthetic origin, reported below.

As mentioned above, the present invention also relates to concentrated solutions comprising at least one viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene in at least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, synthetic

25 origin, or mixtures thereof.

Lubricating base oils of mineral origin derive from well-known oil refining processes such as, for example, distillation, deparaffinization, deasphalting, dearomatization, hydrogenation.

The lubricating base oils of synthetic origin can be preferably selected from:

30 hydrocarbon oils such as, for example, polymerized and hydrogenated terminal or internal olefins; alkylbenzenes; polyphenyls.

A further way of classifying lubricating base oils is the one defined by the

American Petroleum Institute (API) in the publication “Engine Oil Licensing and Certification System” (API EOLCS, 1507 - Industry Services Department, Fourteenth Edition, December 1996, Addendum 1, December 1998).

In accordance with the aforesaid classification method (API), the lubricating base
5 oils are subdivided into five groups according to their chemical-physical and compositional characteristics.

In accordance with the aforesaid classification method (API), the lubricating base oils that can be used in accordance with the present invention, are those belonging to all Groups I, II, III, IV and V, more preferably to Groups III and IV.

10 As mentioned above, the present invention also relates to a composition of lubricating oil comprising at least one viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene, as such or in concentrated solution, and at least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, of synthetic origin, or mixtures
15 thereof.

Accordingly, the present invention also relates to a composition of lubricating oil comprising at least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, of synthetic origin, or mixtures thereof, and at least one viscosity index improver (V.I.I.) additive of lubricating oils comprising a
20 hydrogenated linear polybutadiene, said additive being present in said lubricating composition in an amount comprised between 0.5% by weight and 50% by weight, preferably comprised between 1% by weight and 35% by weight, with respect to the total weight of said composition of lubricating oil.

The aforesaid compositions of lubricating oil can comprise, in addition to said
25 viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene, also other additives capable of improving the viscosity index, detergent additives, dispersing additives, antioxidant additives, friction modifying additives, anti-wear and extreme pressure additives (EP additives), corrosion inhibitors, pour point depressant additives, foam inhibitors, emulsifiers, or mixtures thereof, and
30 the like.

In order to better understand the present invention and to put it into practice, some illustrative and non-limiting examples thereof are reported below.

The analysis and characterization methods reported below were used.

In the case of hydrogenated linear polybutadienes obtained in accordance with the aforesaid second process (i.e. in the presence of at least one lubricating base oil), they must be separated from the oil before being subjected to the analyses reported below.

5 To this end, 500 ml of ethanol (Merck) and a magnetic anchor were added in a 1-liter becker and, subsequently, 25 ml of the hydrogenated linear oil/polybutadiene mixture obtained as reported in the following examples: the solution was kept, under stirring, at room temperature (25 C), for 1 hour. Subsequently, the obtained solution was filtered through a 325-mesh metal filter, obtaining oil-free hydrogenated linear
10 polybutadiene.

Spectra ¹H-NMR

The quantitative determination of the microstructure of the hydrogenated linear polybutadiene in terms of the content of units having 1,2 configuration was carried out with internal method based on ISO 21561-1:2015 standard through ¹H-NMR
15 spectrometry.

¹H-NMR spectra were recorded at 30 C by high resolution liquid phase nuclear magnetic resonance spectrometer mod. Bruker Avance Neo 300 MHz, equipped with 5 mm DUL 13C-1H/2H Z-GRD probe and variable temperature.

For the analyses, polymer solutions of the non-hydrogenated polybutadienes
20 ("parent polymers"), obtained as reported in the following examples, were used, having concentrations equal to 3% w/v (g/ml) prepared using deuterated chloroform (CDCl₃) (purity > 99.8%).

The operating conditions adopted were the following:

Acquisition parameters (5 mm probe):

- 25 - mode: single pulse;
- flip angle (μs): pulse 90 C;
- temperature: 30 C;
- number of data points: 64 k;
- offset: 4.6 ppm;
30 - sweep width: 9 ppm;
- pulse repetition: 10 seconds;
- accumulation: 64;

- solvent: deuterated chloroform (CDCl_3) (Merck: purity > 99.8%);
- sample spinning: yes.

Calculations ^1H :

Integral IA: area of olefinic signals relating to 1,2 polybutadiene units (4.3÷5.0 ppm);

- 5 *Integral IB*: area of the olefinic signals relating to the 1,2 and 1,4 polybutadiene units (5.0÷6.1 ppm)

$$1.2\% = \frac{\frac{IA}{2}}{\frac{IB}{2} + \frac{IA}{4}} \times 100$$

$$1.4\% = \frac{\frac{IB}{2} - \frac{IA}{4}}{\frac{IB}{2} + \frac{IA}{4}} \times 100$$

Molecular weight determination

- The determination of the weight average molecular weight (M_w), the number average molecular weight (M_n), and the polydispersity index (PDI) (M_w/M_n), of the hydrogenated polybutadienes obtained as reported in the following examples, was carried out by GPC (“Gel Permeation Chromatography”), using the integrated 1200 series instrument of Agilent Technologies that uses RID detection, operating under the following conditions:

- 15 - six GPC PL columns of Agilent Technologies having dimensions 300 x 7.5 mm, specifically composed as follows:

TYPE	Particle size	Porosity (Å)
PLGEL	5 μm	10^5
PLGEL	5 μm	10^5
PLGEL	5 μm	10^4
PLGEL	5 μm	10^3
PLGEL	10 μm	10^6
PLGEL	5 μm	500

- 20 - Mettler Toledo analytical balance;
- laboratory glassware;
- shaking stirrer;
- column injection temperature: 25 C;

- column and detector temperature: 25°C;
- solvent/eluent: tetrahydrofuran (THF) (99+% for HPLC - Merck);
- flow: 1 ml/min;
- calculation of molecular mass by universal calibration curve.

5 The aforesaid operating conditions have been constantly monitored through a personal computer equipped with Agilent GPC/SEC software from Agilent Technologies.

Calibration was carried out as follows.

10 Four tetrahydrofuran (THF) solutions (99+% by HPLC - Merck) were prepared each containing three polystyrene (PS) standards having a different nominal peak molecular weight (M_p) and equal concentrations: the selection of molecular weights was made in such a way that the eluted chromatographic peaks are well separated. The range of the different nominal peak molecular weights (M_p) of the aforesaid standards used for the calibration curve was comprised between about 3 kDa and 1200 kDa.

15 The different solutions were prepared, under stirring, at room temperature (25°C).

The calibration curve was calculated using a personal computer equipped with the aforesaid Agilent GPC/SEC software from Agilent Technologies, using a 3rd order polynomial function: through said polynomial it was possible to obtain the aforesaid information relating to the molecular weight.

20 Determination of the degree of hydrogenation (GI)

The determination of the degree of hydrogenation (GI) of the hydrogenated linear polybutadienes obtained as reported in the following examples was carried out by Fourier Transform Infrared Spectroscopy (FTIR), using the NICOLET iS50 spectrophotometer instrument from THERMO FISHER having the following

25 characteristics:

- OMNIC software version 9.12;
- spectral range: 4000-400 cm^{-1} ;
- single beam;
- detector: TGS (triglycine sulphate deuterate).

30 The operating conditions adopted were the following:

- measurement during transmission;
- spectral range: 1050-650 cm^{-1} ;

- number of scans: 25;
- resolution: 4 cm⁻¹;
- gain: 1;
- speed of movement of the mirror in the interferometer: 0.6329 cm/s;
- 5 - opening: 65.

A certain amount of a solution of a hydrogenated linear styrene-butadiene-styrene block copolymer (Europrene® SOL TH 2315) in cyclohexane (Merck) (~10% w/w) was added to each sample to be analysed, operating as reported below.

For the calculation of the calibration line, the non-hydrogenated linear polybutadienes (“parent polymers”) used in the following examples and the hydrogenated linear polybutadienes obtained from said examples were used.

For the aforesaid non-hydrogenated linear polybutadienes (“parent polymers”) it was necessary to determine the absorbencies relative to the functional groups under examination. In detail, it was operated under the following conditions:

- 15 - 0.8 g of non-hydrogenated linear polybutadiene (“parent-polymer”) were weighed in a 20 ml vial with screw cap, then 10 ml of cyclohexane (Merck) (~10% w/w) were added and the whole was placed in a shaking stirrer until completely solubilised;
- 5 ml of the obtained solution were mixed with 5 ml of the solution of a hydrogenated linear styrene-butadiene-styrene block copolymer (Europrene® SOL TH 2315) in cyclohexane (Merck) (10% by weight solution), so as to simulate a styrene-butadiene copolymer with a bound styrene content of around 15% by weight, the obtained mixture was stirred until complete mixing and a thin layer of the obtained polymer solution was spread, with a glass rod, on a rectangular window of potassium bromide (40x20 mm with a thickness of 5 mm) and the solvent was evaporated by a nitrogen flow;
- 20 - a background was made, to be subtracted from the IR spectra of the sample, by carrying out 25 scans in the spectral range comprised between 1050 cm⁻¹ and 650 cm⁻¹, leaving the sample compartment empty;
- 25 - 25 scans of the sample were made in the same spectral field by inserting, along the optical path of the IR ray, the window on which the polymer solution was spread (for a correct evaluation of the spectrum it is necessary that the
- 30

transmittance at 1050 cm⁻¹ is greater than 80%, while the most intense spectral band must be comprised between 20% and 40%; if the conditions described above have not been respected, it is necessary not to consider the value obtained and repeat the analysis of the sample);

- 5 - the maximum absorbance was measured at about 968 cm⁻¹ (At), 910 cm⁻¹ (Av) and 700 cm⁻¹ (As);
- for the “parent polymer” the ratio (At + Av)/As was measured and reported in the graph on the ordinate axis equal to 0; for the totally hydrogenated polymer the ratio (At + Av)/As will be equal to 0; the equation of the linear line $y=aX + b$ that
- 10 interpolates the two points was calculated (reported in Figure 1).

The degree of hydrogenation was calculated by the following expression:

$$GI = a * \frac{Av + At}{As} + b$$

wherein:

- Av = vinyl absorbance (910 cm⁻¹);
- 15 - At = trans absorbance (968 cm⁻¹);
- As = styrene absorbance (700 cm⁻¹);
- a = angular coefficient of the straight line;
- b = Intercept of the straight line on the ordinate axis.

Determination of the enthalpy of crystallization (ΔH_c)

- 20 The determination of the enthalpy of crystallization (ΔH_c) was carried out by means of temperature modulated differential scanning calorimetry (TMDSC) using a Discovery DSC 2500 differential scanning calorimeter from TA Instruments.

- For the theoretical foundations of the temperature modulated differential scanning calorimetry (TMDSC), refer to what is reported by Simon S.L. in “*Thermochimica Acta*”
- 25 (2001), Vol. 374, p. 55-71.

Said temperature modulated differential scanning calorimetry (TMDSC) allows to be able to distinguish processes of the glass transition/melting/crystallization type that occur in the same temperature range according to the equation reported below:

$$\frac{dH}{dt} = Cp \left(\frac{dT}{dt} \right) + f(T, t)$$

- 30 wherein:

- $\frac{dH}{dt}$ is the total heat flow [W/g], H is enthalpy per unit mass [J/g] and t time [s];

- C_p is the specific heat at constant pressure $\left[\frac{J}{g \cdot ^\circ C}\right]$ (heat capacity);
- $\left(\frac{dT}{dt}\right)$ is the heating rate $\left[\frac{^\circ C}{s}\right]$;
- $C_p\left(\frac{dT}{dt}\right)$ is the reversing heat flow [W/g];
- $f(T, t)$ is the non-reversing heat flow [W/g].

5 For this purpose, for the determination of the glass transition temperature (T_g), of the crystallization temperature (T_c) and of the enthalpy of crystallization (ΔH_c), the temperature modulated differential scanning calorimetry (TMDSC) was carried out by applying the following thermal cycle to the samples of the hydrogenated linear polybutadienes obtained as reported in the following examples:

- 10
- sample conditioning: heating from $T = 23 \pm 1$ C up to $T = +100^\circ C$ and subsequent isotherm at $T = +100^\circ C$ for 5 minutes;
 - cooling ramp from $T = +100$ C to $T = -100^\circ C$ at $v = 3^\circ C/min$ in modulation of ± 0.47 C every 60 seconds with TMDSC (i.e. Modulated DSC);

wherein T = temperature; v = scan speed.

15 Determination of the dynamic viscosity of the hydrogenated linear polybutadienes

The dynamic viscosity measurement was carried out with Anton Paar's MCR 702 controlled stress rotational rheometer equipped with steel smooth parallel plate geometry (diameter 40 mm, PP40).

For the theoretical foundations of rheometry and the good practices required for a
20 correct measurement of dynamic viscosity with rotational rheometers equipped with parallel plates, refer to what is reported by Macosko W. in "*Rheology: Principles, Measurements, and Applications*" (1994), Ed. by Wiley-VCH Inc, Chapter 5, pp. 217-220.

The dynamic viscosity measurements were carried out in the temperature range T
25 = 40 C, 80°C, 100°C, 120°C and 150°C. The rheometer software is Rheocompass version 1.31, release 43. The zero-gap procedure (i.e. zeroing of the distance between the plates) was carried out at the temperature $T = 100^\circ C$. The rheometer is able to compensate for the thermal expansion of the measuring accessories caused by the temperature variation. The thermal expansion of the accessories was calibrated using the
30 procedure provided by Anton Paar and available in the Rheocompass software used.

The correct analysis temperature was ensured using a Peltier-type temperature

control system (model C-PTD200 + H-PTD200) equipped with a convection hood. To allow temperature stabilization, a time of 20 minutes was awaited at each analysis temperature.

The viscosity value at steady state was recorded after 100 seconds from the start of the stress application, to exclude the effects due to the start-up viscoelastic transient.

The tests were carried out at different shear stresses set (from 0.1 Pa to 320 Pa) in order to verify the possible non-Newtonian behaviour of the materials. A maximum limit on the shear rate equal to 10 s^{-1} has been imposed in order to avoid problems due to flow instability phenomena.

10 Determination of the target concentration in base oil

For this purpose, the hydrogenated linear polybutadienes obtained were dissolved in a variable amount to obtain the target dynamic viscosity in Group III 4 cSt base oil (Yubase 4 - SK Lubricant) equivalent to the dynamic viscosity, measured in accordance with ASTM D-7042-04 standard by means of the SVM 3000 rotational viscometer from Antoon Paar, of a solution of an ethylene-propylene copolymer (Dutral[®] OCP 2550 - Versalis S.p.A.) at a concentration equal to 1% by weight in the same Group III 4 cSt base oil.

Determination of the shear stability index (SSI)

For this purpose, the hydrogenated linear polybutadienes obtained as reported in the following examples were dissolved in an amount equal to the target concentration in Group III 4 cSt base oil (Yubase 4 - SK Lubricant).

The shear stability index (SSI) was determined in accordance with the CEC-L-14-A-93 standard (30 cycles of shearing).

Determination of the pour point

For this purpose, the hydrogenated linear polybutadienes obtained as reported in the following examples were dissolved in an amount equal to the target concentration in Group III 4 cSt base oil (Yubase 4 - SK Lubricant) and, to the solution obtained, an amount equal to 0.2% by weight with respect to the total weight of said solution of the additive Viscoplex[®] V1-254 (Evonik) ("Pour Point Depressant" - PPD) was added.

The pour point was measured in accordance with ASTM D7346-15 standard.

Determination of the viscosity index

For this purpose, the hydrogenated linear polybutadienes obtained as reported in

the following examples were dissolved in an amount equal to the target concentration in Group III 4 cSt base oil (Yubase 4 - SK Lubricant) and the viscosity index was measured in accordance with ASTM D2270-95 standard.

EXAMPLES 1 AND 3-5 (invention)

5 Preparation of the hydrogenated linear polybutadienes

Four hydrogenated linear polybutadienes were prepared by operating as described below.

The solution polymerization reaction, in discontinuous (batch), was carried out in an 18.5 L batch reactor (of which 16 L are useful), equipped with impeller blades, anchor
10 bottom scraper, bottom discharge and heating jacket.

Said reactor was fed with anhydrous cyclohexane (Cepsa) and, after bringing the temperature to 35 C, tetrahydrofuran (THF) (VWR International) was added and, subsequently, the following reactions were carried out:

Polymerization [(step (a))]

- 15 - addition of anhydrous 1,3-butadiene (Versalis S.p.A.);
- addition of lithium *n*-butyl (15% solution in hexane) (Merck) (NBL);
- waiting for reaction time;

Termination [(step (b))]

- addition of trimethyl chloro silane (CH₃SiCl) (Merck) [step (b)];
- 20 - waiting for reaction time;

Hydrogenation [(step (c))]

- addition of butyl ethyl magnesium chloride (14% solution in heptane) (Nouryon);
- addition of bis-cyclopentadienyl-titanium-dichloride (Nouryon);
- waiting for reaction time.

25 At the end of the hydrogenation step (c), the mixture obtained was subjected to the desolventisation step (d), in the absence of water, by means of a stirred thin film evaporator, operating at a pressure equal to 35 mbar and a temperature equal to 170 C.

Table 1 reports the operating conditions used: feeding the compounds to the reactor, temperature and reaction duration.

30 The hydrogenated linear polybutadienes obtained were subjected to the characterizations reported above and the results obtained are reported in Table 2.

EXAMPLE 2 (invention)

Preparation of the hydrogenated linear polybutadiene in oil

A hydrogenated linear polybutadiene in oil was prepared by operating as described below.

The solution polymerization reaction, in discontinuous (batch), was carried out in
5 a 1 L batch reactor (of which 0.8 L was useful), equipped with impeller blades, bottom discharge and heating jacket.

Said reactor was fed with Group III 4 cSt base oil (Yubase 4 - SK Lubricant) and, after bringing the temperature to 60 C, tetrahydrofuran (THF) (VWR International) was added and, subsequently, the following reactions were carried out:

10 Polymerization [(step (a'))]

- addition of anhydrous 1,3-butadiene (Versalis S.p.A.);
- addition of lithium *n*-butyl (15% solution in hexane) (Merck) (NBL);
- waiting for reaction time;

Termination [(step (b'))]

- 15
- addition of trimethyl chloro silane (CH₃SiCl) (Merck) [step (b')];
 - waiting for reaction time;

Hydrogenation [(step (c'))]

- 20
- addition of butyl ethyl magnesium (14% solution in heptane) (Nouryon);
 - addition of bis-cyclopentadienyl-titanium-dichloride (Nouryon);
 - waiting for reaction time.

Table 1 reports the operating conditions used: feeding the compounds to the reactor, temperature and reaction duration.

The hydrogenated linear polybutadiene obtained was subjected to the characterizations reported above and the results obtained are reported in Table 2.

25 EXAMPLES 6-8 (comparative)

Preparation of the hydrogenated linear polybutadienes

3 hydrogenated linear polybutadienes were prepared by operating as described below.

The solution polymerization reaction, in discontinuous (batch), was carried out in
30 an 18.5 L batch reactor (of which 16 L are useful), equipped with impeller blades, anchor bottom scraper, bottom discharge and heating jacket.

Said reactor was fed with anhydrous cyclohexane (Cepsa) and, after bringing the

temperature to 35 C, tetrahydrofuran (THF) (VWR International) was added and, subsequently, the following reactions were carried out:

Polymerization [(step (a))]

- addition of anhydrous 1,3-butadiene (Versalis S.p.A.);
- 5 - addition of lithium *n*-butyl (15% solution in hexane) (Merck) (NBL);
- waiting for reaction time;

Termination [(step (b))]

- addition of trimethyl chloro silane (CH₃SiCl) (Merck) [step (b)];
- waiting for reaction time;

10 Hydrogenation [(step (c))]

- addition of butyl ethyl magnesium chloride (14% solution in heptane) (Nouryon);
- addition of bis-cyclopentadienyl-titanium-dichloride (Nouryon);
- waiting for reaction time.

At the end of the hydrogenation step (c), the mixture obtained was subjected to
15 the desolventisation step (d), in the presence of water, operating as reported below.

The mixture obtained was discharged from the bottom of the reactor and sent to a storage container. Subsequently, said mixture was sent, continuously, to the strippers filled with water at 90 C, to which steam was also sent, so as to obtain the stripping of the unreacted monomers (1,3-butadiene and styrene) and of the solvent: in fact, it was
20 not possible to carry out desolventisation by means of a thin film evaporator stirred due to the high viscosity of the polymers.

The water leaving the strippers was collected and sent to water treatment.

Table 1 reports the operating conditions used: feeding the compounds to the reactor, temperature and reaction duration.

25 The hydrogenated linear polybutadienes obtained were subjected to the characterizations reported above and the results obtained are reported in Table 2.

Table 1

	Compounds	Example 1 (invention)	Example 2 (invention)	Example 3 (invention)	Example 4 (invention)	Example 5 (invention)	Example 6 (comparative)	Example 7 (comparative)	Example 8 (comparative)
	Cyclohexane (g)	8345	-	8365	8339	8343	8366	8374	8343
	Oil (g)	-	500	-	-	-	-	-	-
	THF (ml)	185	7.3	130	263	225.2	120	24.1	47.5
	THF (ppm with respect to solvent or oil)	12046	12994	13051	26482	22665	12046	2417	4781
Polymerisation [step (a)] [step (a')]]	NBL (15% in <i>n</i> -hexane) (ml)	18.5	1.0	110	49.5	33.8	11.2	17.0	17.5
	NBL (15% in <i>n</i> -hexane) (phm)	0.24	0.51	1.40	0.78	0.43	0.14	0.22	0.22
Termination [step (b)] [step (b')]]	CH ₃ SiCl (ml)	3	0.1	22	8.8	5.4	2.2	3.1	3.1
Operating conditions	T start of Polymerization [step (a)] [step (a')]] (°C)	35	60	35	35	35	35	35	35
	T end of Polymerization [step (b)] [step (a')]] (°C)	80.9	60	56.5	48	80.2	79.4	81.8	81.2
	Polymerization Duration [step (a)] [step (a')]] (min)	30	30	30	30	30	30	30	30

	T end of Termination [step (b)] [step (b')]] (°C)	80.9	60	56.5	48	80.2	79.4	81.8	81.2
	Termination Duration [step (b)] [step (b')]] (min)	10	10	10	10	10	10	10	10
Hydrogenation [step (c)] [step (c')]]	titanium (calculated as metallic Ti) (ppm)	350	250	300	300	250	350	300	250
	Molar ratio Mg/Ti (mol/mol)	3	11	3	3	3	3	3	3
	T start of hydrogenation [step (c)] [step (c')]] (°C)	96.4	120	95	97.7	99	108	98	95
	Hydrogenation Duration [step (c)] [step (c')]] (min)	45	60	135	140	110	95	80	40

Table 2

Characteristics	Example 1 (invention)	Example 2 (invention)	Example 3 (invention)	Example 4 (invention)	Example 5 (invention)	Example 6 (comparative)	Example 7 (comparative)	Example 8 (comparative)
M _w (Da)	33900	36900	5200	10600	20700	54300	34800	35500
PDI (M _w /M _n)	1.04	1.04	1.03	1.03	1.02	1.02	1.02	1.02
1.2 ⁽¹⁾ (% by weight)	54.5	53.5	59.8	71.3	68	58.7	32.3	41
Degree of hydrogenation (% by weight)	> 99	> 99	> 99	> 99	> 99	> 99	> 99	> 99
Enthalpy of crystallization (ΔH _c) (J/g)	0	0	0	0.4	0	0.7	13	13
Dynamic viscosity (measured at 40 °C)	6390	6870	14.89	145.19	552	45500	N/A ⁽²⁾	14700
Dynamic viscosity (measured at 100 °C) (Pa*s)	136	147	0.57	3.5	12.5	1180.1	466	351.1
Target concentration in oil (% by weight)	2.6	2.2	9.5	5.7	3.9	1.9	1.8	2
Target dynamic viscosity (measured at 100 °C) (cSt)	7.68	7.65	7.87	7.50	7.47	7.53	7.70	7.75
Pour Point (°C)	-39	-39	-36	-36	-39	-36	-30	-24
SSI (30 cycles of shearing)	1	2	1	1	1	4	6	3

- ⁽¹⁾: % by weight with respect to the total weight of the butadiene units in the hydrogenated linear polybutadiene [% by weight determined on the non-hydrogenated linear polybutadiene ("parent polymer")];
- ⁽²⁾: not determined as the sample, at 40°C, was in semi-crystalline state.

CLAIMS

1. Viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene, said hydrogenated linear polybutadiene having the following characteristics:
- 5
- weight average molecular weight (M_w) comprised between 1000 Dalton and 40000 Dalton, preferably comprised between 1500 Dalton and 38000 Dalton;
 - a content of units having configuration 1,2 comprised between 40% by weight and 80% by weight, preferably comprised between 45% by weight and 75% by weight,
 - 10 with respect to the total amount of butadiene units present in said hydrogenated linear polybutadiene;
 - a polydispersion index (PDI), that is, the ratio between the weight average molecular weight (M_w) and the number average molecular weight (M_n) (M_w/M_n), less than or equal to 1.3, preferably comprised between 1 and 1.1;
 - 15 - a degree of hydrogenation greater than or equal to 98%, preferably comprised between 98.5% and 100%;
 - a dynamic viscosity, measured at 40°C, comprised between 5 Pa*s and 10000 Pa*s, preferably comprised between 10 Pa*s and 8000 Pa*s;
 - a dynamic viscosity, measured at 100 C, comprised between 0.1 Pa*s and 1000
 - 20 Pa*s, preferably comprised between 0.2 Pa*s and 500 Pa*s;
 - an enthalpy of crystallization (ΔH_c), determined by temperature modulated differential scanning calorimetry (TMDSC), comprised between 0 J/g and 1 J/g, preferably comprised between 0 J/g and 0.8 J/g.
2. Viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene according to claim 1, having the following characteristics:
- 25
- a shear stability index (SSI) comprised between 0% and 7%, preferably comprised between 0% and 5%, measured in accordance with the CEC-L-14-A-93 standard (30 cycles of shearing) in Group III 4 cSt base oil;
 - 30 - a pour point lower than or equal to -30°C, preferably comprised between -33°C and -45 C, measured in accordance with the ASTM D5950-14 standard in Group

III 4 cSt base oil in the presence of the additive Viscoplex[®] V1-254 (Evonik) (Pour Point Depressant - PPD).

3. Viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene according to claim 1 or 2, wherein said viscosity index improver additive (V.I.I.) of lubricating oils in order to reach a target dynamic viscosity measured in accordance with the ASTM D7042-04 standard, at 100°C, in Group III 4 cSt base oil comprised between 6 cSt and 9 cSt, preferably comprised between 7 cSt and 8 cSt, is used in said Group III 4 cSt base oil at a target concentration comprised between 1.9% by weight and 12% by weight, preferably comprised between 2.1% by weight and 10% by weight, with respect to the total weight of the Group III 4 cSt base oil and viscosity index improver (V.I.I.) of lubricating oils composition.
4. Discontinuous (batch) process for the preparation of a hydrogenated linear polybutadiene comprising the following steps:
 - (a) subjecting 1,3-butadiene to living polymerization by an anionic route in the presence of at least one hydrocarbon solvent, at least one lithium-based initiator and at least one polar modifier and continuing said polymerization until substantially complete conversion of the 1,3-butadiene;
 - (b) adding at least one termination agent to the polymerization mixture obtained in step (a);
 - (c) subjecting the linear polybutadiene obtained in step (b) to hydrogenation obtaining a mixture comprising hydrogenated linear polybutadiene;
 - (d) subjecting the mixture comprising hydrogenated linear polybutadiene obtained in step (c) to desolventisation and recovering the hydrogenated linear polybutadiene;wherein said desolventisation step (d) is carried out in the absence of water.
5. Discontinuous (batch) process for the preparation of a hydrogenated linear polybutadiene according to claim 4, wherein:
 - said at least one hydrocarbon solvent is selected from aliphatic, cycloaliphatic or aromatic hydrocarbon solvents such as propane, *n*-butane, *iso*-butane, *n*-pentane, *iso*-pentane, *n*-hexane, *n*-heptane, *n*-octane, cyclohexane, cyclopentane, propene, 1-butene, *iso*-butene, *trans*-2-butene, *cis*-2-butene, 1-pentene, 2-pentene, 1-hexene, 2-hexene, benzene, toluene, xylene, ethylbenzene, or mixtures thereof,

preferably in anhydrous form; it is preferably selected from cyclohexane, *n*-hexane, or mixtures thereof, in anhydrous form; and/or

- said at least one lithium-based initiator is selected from compounds having a general formula (I):



wherein R_1 represents an alkyl group C_1 - C_{20} , preferably C_2 - C_{12} , linear or branched, a cycloalkyl group C_3 - C_{30} , preferably C_4 - C_{10} , an aryl group C_6 - C_{30} , preferably C_6 - C_{12} , and n is an integer comprised between 1 and 4; preferably between: lithium methyl, lithium *n*-butyl, lithium *sec*-butyl, lithium *t*-butyl, lithium *n*-propyl, lithium *iso*-butyl, lithium amyl, lithium cyclohexyl, lithium phenyl, lithium 1-methyl styryl, lithium *p*-tolyl, lithium naphthyl, L,L-diphenyl-5-lithium-3-methyl-pentyl, or mixtures thereof; more preferably it is lithium *n*-butyl; and/or

- said at least one polar modifier is selected from: non-cyclic ethers such as ethyl ether, or mixtures thereof; tertiary amines such as tri-butyl amine; cyclic ethers such as tetrahydrofuran (THF); chelating ethers such as ethylene glycol dimethyl ether (dimethylglyme), dioxane, 2-methoxy-ethyl-tetrahydrofuran (THFA-ethyl), 2-methoxy ethyl-tetrahydropyran, or mixtures thereof; chelating amines such as *N,N,N',N'*-tetramethyleneethylenediamine (TMEDA); or mixtures thereof; preferably it is selected from tetrahydrofuran (THF), 2-methoxy-ethyl-tetrahydrofuran (THFA-ethyl), or mixtures thereof; and/or
- said at least one termination agent is selected from trimethyl chlorosilane, butanol, octanol, or mixtures thereof; preferably it is trimethyl chlorosilane.

6. Discontinuous (batch) process for the preparation of a hydrogenated linear polybutadiene according to claim 4 or 5, wherein:

- said step (a) is carried out at a temperature comprised between 20°C and 100°C, preferably comprised between 25°C and 85°C; and/or for a time comprised between 10 minutes and 1 hour, preferably comprised between 15 minutes and 40 minutes; and/or
- said step (b) is carried out at a temperature comprised between 30°C and 115°C, preferably comprised between 40°C and 110°C, and/or for a time comprised between 1 minute and 30 minutes, preferably comprised between 3 minutes and

20 minutes.

7. Discontinuous (“batch”) process, by an anionic route, for the preparation of a hydrogenated linear polybutadiene comprising the following steps:

- 5 (a') subjecting 1,3-butadiene to living polymerization by an anionic route in the presence of at least one lubricating base oil, at least one lithium-based initiator and at least one polar modifier and continuing said polymerization until substantially complete conversion of the 1,3-butadiene;
- (b') adding at least one termination agent to the polymerization mixture obtained in step (a');
- 10 (c') subjecting the linear polybutadiene obtained in step (b') to hydrogenation, obtaining a mixture comprising hydrogenated linear polybutadiene.

8. Discontinuous (“batch”) process, by an anionic route, for the preparation of a hydrogenated linear polybutadiene according to claim 7, wherein:

- said step (a') is carried out at a temperature comprised between 20 C and 100°C, preferably comprised between 25°C and 85°C; and/or for a time comprised between 10 minutes and 1 hour, preferably comprised between 20 minutes and 40 minutes; and/or
- 15 - said step (b') is carried out at a temperature comprised between 30 C and 115 C, preferably comprised between 40 C and 110°C; and/or for a time comprised between 1 minute and 30 minutes, preferably comprised between 3 minutes and 20 minutes.

9. Discontinuous (“batch”) process, by an anionic route, for the preparation of a hydrogenated linear polybutadiene according to claim 7 or 8, wherein:

- said at least one lithium-based initiator, at least one polar modifier, and at least one termination agent, are selected from those referred to in claim 5; and/or
- 25 - said at least one lubricating base oil is selected from lubricating base oils of mineral origin, of synthetic origin, or mixtures thereof.

10. Concentrated solutions comprising at least one viscosity index improver (V.I.I.) additive of lubricating oils comprising a hydrogenated linear polybutadiene according to any one of the previous claims, in at least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, synthetic origin, or
- 30

mixtures thereof.

11. Composition of lubricating oil comprising at least one lubricating base oil, said lubricating base oil being selected from lubricating base oils of mineral origin, of synthetic origin, or mixtures thereof, and at least one viscosity index improver (V.I.I.)
- 5 additive of lubricating oils comprising a hydrogenated linear polybutadiene according to any one of claims 1 to 9, said additive being present in said lubricating composition in an amount comprised between 0.5% by weight and 50% by weight, preferably comprised between 1% by weight and 35% by weight, with respect to the total weight of said lubricating oil composition.