TRIBOTECHNICAL LUBRICANT AND LUBRICANT COMPOSITION

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Appl. No.: 12/026,725
Filed: Feb. 6, 2008

Foreign Application Priority Data
Feb. 6, 2007 (RU) .............................. 2007105032

Abstract

The tribotechnical lubricant and lubricant composition may be used for friction decrease in friction units of machinery and equipments included heavy-loaded ones in aging and operation process. The tribotechnical lubricant contains x-ray amorphous carbon characterized by particle size by electron microscopy of 30-50 nm, temperature of oxidation beginning in air is not more than 300°C, nonconjugated double bonds concentration determined on interaction with neutral potassium permanganate solution is not less than one per 5 carbon atoms, paramagnetic centers concentration on ESR-line with g factor 2.022 is not less than one per 1200 carbon atoms. The lubricant composition contains a base oil and x-ray amorphous carbon.
Tribotechnical lubricant and lubricant composition

Table 1

<table>
<thead>
<tr>
<th>[AC], μg · mm⁻²</th>
<th>Constant of friction on steady-state wear stage</th>
<th>The linear wear, μm</th>
<th>The linear wear intensity</th>
<th>( I_b \cdot 10^{10} )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 MPa</td>
<td>3 MPa</td>
<td>5 MPa</td>
<td>1 MPa</td>
</tr>
<tr>
<td>-</td>
<td>0.49-0.50</td>
<td>0.57-0.59</td>
<td>0.58-0.63</td>
<td>3.2</td>
</tr>
<tr>
<td>0.5</td>
<td>0.48</td>
<td>0.56</td>
<td>0.61</td>
<td>2.3</td>
</tr>
<tr>
<td>2.5</td>
<td>0.46</td>
<td>-</td>
<td>0.58</td>
<td>0.7</td>
</tr>
<tr>
<td>3.75</td>
<td>0.37</td>
<td>0.50-0.51</td>
<td>0.54</td>
<td>0.9</td>
</tr>
<tr>
<td>5.0</td>
<td>0.46-0.47</td>
<td>0.51-0.52</td>
<td>0.61</td>
<td>3.6</td>
</tr>
</tbody>
</table>

Fig. 2

Table 2

<table>
<thead>
<tr>
<th>[AC], μg · mm⁻²</th>
<th>The linear wear, μm</th>
<th>The linear wear intensity</th>
<th>( I_b \cdot 10^{10} )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 MPa</td>
<td>3 MPa</td>
<td>5 MPa</td>
</tr>
<tr>
<td>-</td>
<td>1.0</td>
<td>1.6</td>
<td>1.2</td>
</tr>
<tr>
<td>0.1</td>
<td>1.5</td>
<td>-</td>
<td>1.3</td>
</tr>
<tr>
<td>2.5</td>
<td>1.7</td>
<td>1.4</td>
<td>14.0</td>
</tr>
<tr>
<td>3.75</td>
<td>0.8</td>
<td>-</td>
<td>8.3</td>
</tr>
<tr>
<td>5.0</td>
<td>0.2</td>
<td>1.7</td>
<td>2.1</td>
</tr>
</tbody>
</table>
Tribotechnical lubricant and lubricant composition

Fig. 3

Fig. 4
Tribotechnical lubricant and lubricant composition

**Fig. 5**

**Fig. 6**
Tribotechnical lubricant and lubricant composition

Fig. 7

Fig. 8
Tribotechnical lubricant and lubricant composition

Fig. 9

Fig. 10
Tribotechnical lubricant and lubricant composition

Fig. 11

Fig. 12
Tribotechnical lubricant and lubricant composition

Table 3

<table>
<thead>
<tr>
<th>[AC], wt. %</th>
<th>Constant of friction</th>
<th>The linear wear, μm</th>
<th>The linear wear intensity $J_h \cdot 10^{11}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>of 45 steel</td>
<td>of a rider</td>
</tr>
<tr>
<td>-</td>
<td>0.12-0.13</td>
<td>1.8</td>
<td>0.4</td>
</tr>
<tr>
<td>0.1</td>
<td>0.12-0.12</td>
<td>1.4</td>
<td>0.4</td>
</tr>
<tr>
<td>0.5</td>
<td>0.11-0.11</td>
<td>0.6</td>
<td>0.4</td>
</tr>
<tr>
<td>3.0</td>
<td>0.09-0.10</td>
<td>0.6</td>
<td>0.1</td>
</tr>
<tr>
<td>5.0</td>
<td>0.09</td>
<td>0.6</td>
<td>0.1</td>
</tr>
<tr>
<td>7.0</td>
<td>0.08-0.09</td>
<td>0.6</td>
<td>0.9</td>
</tr>
</tbody>
</table>

Fig. 13

![Figure 13](image)

Fig. 14

![Figure 14](image)
Tribotechnical lubricant and lubricant composition
Tribotechnical lubricant and lubricant composition

Fig. 17

Fig. 18
Tribotechnical lubricant and lubricant composition

Fig. 19

Fig. 20
Tribotechnical lubricant and lubricant composition

Fig. 21

Fig. 22
Tribotechnical lubricant and lubricant composition

Fig. 23

Fig. 24
Tribotechnical lubricant and lubricant composition

Fig. 25
TRIBOTECHNICAL LUBRICANT AND LUBRICANT COMPOSITION

RELATED APPLICATIONS

[0001] This application claims priority to Russian Patent Application No. RU 2007105032 filed on Feb. 6, 2007, which is incorporated herein by reference in its entirety.

FIELD OF THE INVENTION

[0002] The present invention relates to petroleum refining, oil and grease production and to lubricant composition destined for reducing friction in friction units of machinery and equipments included heavy-loaded ones in aging and operation process.

BACKGROUND OF THE INVENTION

[0003] A semisolid lubricant (RU2268291, published 20 Jan. 2006) containing base viscous lubrication and carbon additive such as 1-5 wt. % of powder of fullerene-containing soot or its powder after fullerene extraction include incompatible hydrated lime soaps and hydrophobic soot as a result of this homogeneous lubricant can not be produced.

[0004] Also a lubricant (U.S. Pat. No. 5,958,523, published 28 Sep. 1999) as product of interaction of fullerene and polytetrafluoroethylene as a whole or in aprotic solvent media at fluorescent irradiation is known. The fullerene lubricant (US20050221995, published 6 Oct. 2005) inclusive free fullerenes or mixture of fullerenes and oil is known also.

[0005] A lubrication-cleaning composition (RU No 2217481, published 27 Nov. 2003) inclusive organic base as mixture of liquid paraffin with chlorine content of 45-47 wt. % stabilized by 0.9-1.1 wt. % of epoxy-diane rubber and filling agent as fullerene schungite C_{60},C_{70} crushed to particle size of 100-500 μm and adipinic acid ester, and antioxidant such as 2,2,4-trimethyl-1,2-dihydroquinoline oligomer and lightstabilizer is known.

[0006] A known lubrication-cleaning composition has complicated formula inclusive abrasive schungite that is impossible crushed to pseudo-molecular particle size.

[0007] A lubricating composition (JP2005054008, published 3 Mar. 2005) inclusive base oil and diurea compound as thickener and carbon powder with mean diameter of particles of ≤1.000 μm such as carbon black or carbon nanotubes or fullerene is known. A lubricating composition (U.S. Pat. No. 5,292,444, published 8 Mar. 1994) inclusive base oil and interaction product of fullerenes C_{60},C_{70},C_{75},C_{80},C_{84},C_{120} or its mixture and amino polymer compound and other additives as dispersant, antioxidant etc. is known. A such composition used high-priced additives, some from which is unstable in operational environment.

SUMMARY OF THE INVENTION

[0011] In one aspect of the invention, x-ray amorphous carbon what is characterized by particle size by electron microscopy of 30-50 nm;
[0012] temperature of oxidation beginning in air is not more than 300° C.;
[0013] nonconjugated double bonds concentration determined on interaction with neutral potassium permanganate solution is not less than one per 5 carbon atoms;
[0014] paramagnetic centers concentration on ESR-line with g-factor 2.022 is not less than one per 1200 carbon atoms used as tribological lubricant.

[0015] The surface concentration of this tribological lubricant may be equal of 0.5-5.0 μg/mm².

[0016] In a second aspect of the invention, in friction zone introduced lubrication composition inclusive base oil and x-ray amorphous carbon what is characterized by particle size by electron microscopy of 30-50 nm;
[0017] temperature of oxidation beginning in air is not more than 300° C.;
[0018] nonconjugated double bonds concentration determined on interaction with neutral potassium permanganate solution is not less than one per 5 carbon atoms;
[0019] paramagnetic centers concentration on ESR-line with g-factor 2.022 is not less than one per 1200 carbon atoms;
[0020] at following proportion (in wt. %);
[0021] x-ray amorphous carbon 0.5-7.0;
[0022] base oil in the rest.

[0023] Fullerene and its crystal form as fullerite, pyrocarbon and active carbon not conform to x-ray amorphous carbon characteristics since its are not x-ray amorphous, its temperatures of oxidation beginning in air is more than 300° C., its have not paramagnetic centers concentration on ESR-line with g-factor 2.022 less than one per 1200 carbon atoms.

[0024] The totality of characteristics fullerene soot and amorphous carbon by patent RU2277557 is not conformed to ones for x-ray amorphous carbon as its temperatures of oxidation beginning in air is more than 300° C., its have not paramagnetic centers concentration on ESR-line with g-factor 2.022 less than one per 1200 carbon atoms.

[0025] X-ray amorphous carbon may be produced by electron or laser evaporation of carbonic material. To this end...
carbonic material is vaporized in helium atmosphere at supply to it energy flow equal 50-300 Wt-mm$^2$, deposition of fullerene soot formed at evaporation, extraction of fullerenes from named soot by organic solvent, separation of precipitate, its washing and drying. The said drying carried out in vacuum at temperature 150-200$^\circ$ C.

[0026] Graphite can be used in claimed method as carbonic material.

[0027] The evaporation of carbonic material in helium atmosphere can be carried out by electric arc discharge with energy flow 50-300 Wt-mm$^2$ produced in cylindrical chamber with coaxial electrodes at proportion of chamber diameter to electrode diameter equal (10-20):1. At least one from electrodes can be made from graphite. In this case on the made from graphite electrode applies voltage with positive polarity and this electrode is shifted towards opposite electrode with the shift rate 0.2-6.0 mm/min.

[0028] Evaporation of carbonic material carries out mainly at helium pressure 100-760 Torr.

[0029] X-ray amorphous carbon may be produced also by pyrolysis of a mixture of hydrogen and hydrocarbon selected from alkanes C$_m$, alkenes C$_m$, alkynes C$_m$, cycloalkanes C$_m$, or its mixture at volume ratio equals (0.1-7):1 over nickel or ferrous catalyst at temperatures of 500-800$^\circ$ C and following isolation of x-ray amorphous carbon produced by magnetic separation.

[0030] All criteria mentioned are necessary for production of the claimed tribotechnical lubricant and lubricant composition. Without catalyst or at temperatures less than 500$^\circ$ C, hydrocarbons of feed are not converted. At pyrolysis temperatures more than 800$^\circ$ C or at use of other not named hydrocarbons inactive materials contained pyrocarbon are formed. A dilution of hydrocarbons by hydrogen superimposed to it formed at pyrolysis inhibits graphite formation must by means of its hydrogenolysis.

[0031] A pyrolysis of named mixture of hydrocarbon and hydrogen over nickel or ferrous catalyst at temperatures of 500-800$^\circ$ C may be realized in flow reactor at feed gas space velocity of 100-10000 hr$^{-1}$ or in static conditions.

[0032] X-ray amorphous carbon have specific surface area of 210-280 m$^2$g$^{-1}$ and bulk density of $\rho$=0.05 g/cm$^3$.

[0033] Hydrogen gas jettable from solid x-ray amorphous carbon is useful by-product of pyrolysis of named mixture of hydrogen and hydrocarbon that is one from merits of this method of x-ray amorphous carbon production.

[0034] The above and other features of the invention including various novel details of construction and combinations of parts, and other advantages, will now be more particularly described with reference to the accompanying drawings and pointed out in the claims. It will be understood that the particular method and device embodying the invention are shown by way of illustration and not as a limitation of the invention. The principles and features of this invention may be employed in various and numerous embodiments without departing from the scope of the invention.

**BRIEF DESCRIPTION OF THE DRAWINGS**

[0035] In the accompanying drawings, reference characters refer to the same parts throughout the different views. The drawings are not necessarily to scale; emphasis has instead been placed upon illustrating the principles of the invention. Of the drawings:

[0036] on FIG. 1 and in table 1 are adduced the tribotechnical characteristics of pair of 45 steel/65G steel at the friction at different surface concentration of tribotechnical lubricant as x-ray amorphous carbon;

[0037] on FIG. 2 and table 2 are adduced linear wear and wear-rate of rider (65 G steel) at the friction at different surface concentration of tribotechnical lubricant (x-ray amorphous carbon);

[0038] on FIG. 3 is adduced sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 1 MPa (1), 3 MPa (2), 5 MPa (3) without tribotechnical lubricant (x-ray amorphous carbon);

[0039] on FIG. 4 is adduced sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 1 MPa (1), 3 MPa (2) at tribotechnical lubricant surface concentration $[\text{AC}]$=2.5 $\mu$gmm$^{-2}$;

[0040] FIG. 5 is adduced sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 1 MPa (1), 3 MPa (2) and 5 MPa at tribotechnical lubricant surface concentration $[\text{AC}]$=3.75 $\mu$gmm$^{-2}$;

[0041] on FIG. 6 is adduced sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 1 MPa (1), 3 MPa (2) and 5 MPa at tribotechnical lubricant surface concentration of $[\text{AC}]$=5.0 $\mu$gmm$^{-2}$;

[0042] on FIG. 7 surface concentration of tribotechnical lubricant $[\text{AC}]$ in friction zone relations of constant of friction at contact pressure $\rho$ equal 1 MPa (1), 3 MPa (2) and 5 MPa are shown;

[0043] on FIG. 8 sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 10 MPa at lubrication by base oil without x-ray amorphous carbon is shown;

[0044] on FIG. 9 sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 10 MPa at lubrication by lubricant composition with x-ray amorphous carbon content of 0.5 wt. % is shown;

[0045] on FIG. 10 sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 10 MPa at lubrication by lubricant composition with x-ray amorphous carbon content of 3.0 wt. % is shown;

[0046] on FIG. 11 sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 10 MPa at lubrication by lubricant composition with x-ray amorphous carbon content of 5.0 wt. % is shown;

[0047] on FIG. 12 sliding distance relation of constant of friction of 45 steel at contact pressure $\rho$ equal 10 MPa at lubrication by lubricant composition with x-ray amorphous carbon content of 7.0 wt. % is shown;

[0048] on FIG. 13 in table 3 linear wear and wear-rate of sample (45 steel) and rider (65 G steel) are shown for friction without base oil and at different x-ray amorphous carbon content;

[0049] on FIG. 14 x-ray amorphous carbon content relation of constant of friction at contact pressure $\rho$ equal 10 MPa is shown;

[0050] on FIG. 15 micrograph of friction surface structure of samples without lubricant at contact pressure $\rho$ equal 3 MPa is shown;

[0051] on FIG. 16 micrograph of friction surface structure of samples at x-ray amorphous carbon surface concentration $[\text{AC}]$=3.75 $\mu$gmm$^{-2}$ at contact pressure $\rho$ equal 3 MPa is shown.
[0052] on FIG. 17 micrograph of friction surface structure of samples at X-ray amorphous carbon surface concentration [AC] = 5.00 µg·mm⁻² at contact pressure p equal 3 MPa is shown;

[0053] on FIG. 18 micrograph of friction surface structure of rider without X-ray amorphous carbon at contact pressure p equal 3 MPa is shown;

[0054] on FIG. 19 micrograph of friction surface structure of samples at X-ray amorphous carbon surface concentration [AC] = 3.75 µg·mm⁻² at contact pressure p equal 3 MPa is shown;

[0055] on FIG. 20 micrograph of friction surface structure of samples at X-ray amorphous carbon surface concentration [AC] = 5.00 µg·mm⁻² at contact pressure p equal 3 MPa is shown;

[0056] on FIG. 21 micrograph of friction surface structure of sample at lubrication by base oil without X-ray amorphous carbon at contact pressure p equal 10 MPa is shown;

[0057] on FIG. 22 micrograph of friction surface structure of sample at lubrication by lubricant composition with X-ray amorphous carbon concentration equal 0.5% wt. at contact pressure p equal 10 MPa is shown;

[0058] on FIG. 23 micrograph of friction surface structure of sample at lubrication by lubricant composition with X-ray amorphous carbon concentration equal 3.0% wt. at contact pressure p equal 10 MPa is shown;

[0059] on FIG. 24 micrograph of friction surface structure of sample at lubrication by lubricant composition with X-ray amorphous carbon concentration equal 5.0% wt. at contact pressure p equal 10 MPa is shown;

[0060] on FIG. 25 micrograph of friction surface structure of sample at lubrication by lubricant composition with X-ray amorphous carbon concentration equal 7.0% wt. at contact pressure p equal 10 MPa is shown.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0061] A tribotechnical test of both tribotechnical lubricant and lubricant composition are carried out by a device providing of rider back-and-forth motion with respect to stationary sample. A tested sample in disk form with friction surface as plane-parallel plate with size of 5x8 mm² on it are made from 45 steel. A rider as platen with size of 90x22 mm² thick of 5 mm is made from 65 G steel. A friction surfaces of both sample and rider are polished to roughness Rₐ, equal of 0.32-0.35 µm.

[0062] A rider slip velocity is equal of 0.1 m·sec⁻¹. The E-40A oil is used as base oil. A wear of samples are measured by gravimetric method with following recalculating to a linear wear. An intensity of linear wear is calculated as Iₓ = h/L wherein h is thickness of thickness frayed layer in length units on sliding distance L.

[0063] A friction of 45 steel on 65 G steel is accompanied by intensive adhesive contact of both materials of this tribological situation. A streaky micrelief with gap traces on the friction surface of 45 steel are formed as on FIGS. 14-20 are shown. The average value of intensity of linear wear at p=1 MPa is Iₓ = 2.2·10⁻⁶. A steel microhardness is increase and equal of 5900 MPa. The constant of friction on initial stages of aging is increase but it is constant and equal p=0.45-0.47 on steady-state wear stage as FIGS. 4-6 are shown. A like situation is observed when x-ray amorphous carbon in friction zone is introduced at surface concentration of 0.5, 2.5 or 5.0 µg·mm⁻². The introducing of x-ray amorphous carbon at surface concentration of 3.75 µg·mm⁻² leads to sharp decrease of constant of friction to level of p=0.37-0.40.

[0064] The use of lubricant composition decreases of aging stage as FIGS. 8-14 are shown. The increase of X-ray amorphous carbon concentration in base oil leads to the constant of friction decrease (as FIGS. 13-14 are shown), to formation of smoothed friction surface as on FIG. 25 with high level of microhardness as 5900-6400 MPa and low intensity of linear wear as Iₓ = 5.1·10⁻¹⁰ that points to formation of wearproof structures on friction surface as on FIGS. 13-14, 21-25 are shown.

[0065] A claimed lubricant composition excels in a stability to sedimentation at a storage. The technical effect observed of high stability to sedimentation is concerned with small particle sizes and with interaction of nonconjugated double bonds of x-ray amorphous carbon with composition base oil.

[0066] X-ray amorphous carbon hydrophobicity allows of lubricant use in friction units exploitable at water contact, for example in water transfer pumps or in gravel pumps.

[0067] While this invention has been particularly shown and described with references to preferred embodiments thereof, it will be understood by those skilled in the art that various changes in form and details may be made therein without departing from the scope of the invention encompassed by the appended claims.

What is claimed is:

1. The tribotechnical lubricant in a form of solid carbon material wherein x-ray amorphous carbon characterized by particle size by electron microscopy of 30-50 nm; temperature of oxidation beginning in air is not more than 300° C.; nonconjugated double bonds concentration determined on interaction with neutral potassium permanganate solution is not less than one per 5 carbon atoms; paramagnetic centers concentration on ESR-line with g-factor 2.022 is not less than one per 1200 carbon atoms, is used as solid carbon material.

2. The tribotechnical lubricant on claim 1 wherein its concentration in the friction zone is 0.5-5.0 µg·mm²;

3. The tribotechnical lubricant on claim 1 wherein named x-ray amorphous carbon is produced by evaporation of carbonic material.

4. The tribotechnical lubricant on claim 3 wherein an evaporation of carbonic material is produced by laser emission.

5. The tribotechnical lubricant on claim 3 wherein an evaporation of carbonic material is produced by electric arc.

6. The tribotechnical lubricant on claim 3 wherein named x-ray amorphous carbon is produced by pyrolysis of mixture of hydrogen and hydrocarbon selected from group contained of Cₓ, alkanes, Cₓ, alkenes, Cₓ, alkynes or Cₓ, cycloalkanes with volume ratio of hydrogen/hydrocarbon equals of (0.1-7):1 over nickel or ferrous catalyst at temperatures of 500-800° C. and subsequent segregation of obtained x-ray amorphous carbon from catalyst by magnetic separation.

7. The tribotechnical lubricant on claim 3 wherein named x-ray amorphous carbon has specific surface S=210-280 m²·g⁻¹ and bulk density ρ≤0.05 g·cm⁻³.

8. The lubricant composition inclusive base oil and solid carbon material wherein x-ray amorphous carbon characterized by:

   particle size by electron microscopy of 30-50 nm; temperature of oxidation beginning in air is not more than 300° C.;
nonconjugated double bonds concentration determined on interaction with neutral potassium permanganate solution is not less than one per 5 carbon atoms; paramagnetic centers concentration on ESR-line with g-factor 2.022 is not less than one per 1200 carbon atoms, is used as solid carbon material at following ingredients ratio (in wt. %):
x-ray amorphous carbon—0.5-7.0;
base oil—in the rest.
9. The lubricant composition on claim 8 wherein named x-ray amorphous carbon is produced by evaporation of carbonic material.
10. The lubricant composition on claim 9 wherein an evaporation of carbonic material is produced by laser emission.

11. The lubricant composition on claim 9 wherein an evaporation of carbonic material is produced by electric arc.
12. The lubricant composition on claim 8 wherein named x-ray amorphous carbon is produced by pyrolysis of mixture of hydrogen and hydrocarbon selected from group contained of C₄₊ alkanes, C₂₋₃ alkenes, C₂₋₃ alkydes or C₂₋₃ cycloalkanes with volume ratio of hydrogen/hydrocarbon equals of (0.1-7):1 over nickel or ferrous catalyst at temperatures of 500-800°C, and subsequent segregation of obtained x-ray amorphous carbon from catalyst by magnetic separation.
13. The lubricant composition on claim 8 wherein named x-ray amorphous carbon has specific surface S=210-280 m²/g and bulk density ρ≤0.05 g·cm⁻³.