# 

# (11) EP 3 018 192 A1

(12)

# **EUROPEAN PATENT APPLICATION**

(43) Date of publication:

11.05.2016 Bulletin 2016/19

(51) Int Cl.:

C10M 169/02 (2006.01)

(21) Application number: 15192511.2

(22) Date of filing: 02.11.2015

(84) Designated Contracting States:

AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR

Designated Extension States:

**BA ME** 

**Designated Validation States:** 

MA MD

(30) Priority: 05.11.2014 JP 2014225309

(71) Applicant: Nippon Grease Co., Ltd. Osaka-shi, Osaka 542-0081 (JP)

(72) Inventors:

 IWAMATSU, Hiroki Kobe-shi, Hyogo 653-0055 (JP)

 INADA, Takehito Kobe-shi, Hyogo 653-0055 (JP)

(74) Representative: TBK
Bavariaring 4-6
80336 München (DE)

#### (54) BIODEGRADABLE GREASE COMPOSITION FOR AEROGENERATOR

(57) An object of the present invention is to provide a biodegradable grease composition which is excellent in fretting resistance, extreme pressure property, low temperature property, and biodegradability as well, and has a small effect to the environment even if released to a natural environment. The grease composition comprises a base oil composed of an ester oil having a kinematic viscosity at 40°C of 60 to 160 mm²/s and a thickener

composed of a diurea compound obtained by allowing an amine mixture comprising a 4-8C alicyclic monoamine and a 20-24C aliphatic monoamine to react with a diisocyanate compound, wherein a content molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture is 7:3 to 9:1, the content of the thickener in a total amount of the base oil and the thickener is 7 to 11 % by mass and a penetration is 265 to 340.

#### Description

10

15

20

30

35

40

50

55

#### **TECHNICAL FIELD**

<sup>5</sup> **[0001]** The present invention relates to a biodegradable grease composition for an aerogenerator.

#### BACKGROUND OF THE INVENTION

**[0002]** In recent years, protection of the global environment has been a problem to be solved in all industries. The environment under which a grease composition is used is mostly a closed system and thus an effect on a natural environment is considered to be small. However, for unintended outflow into a natural world due to an accident or leakage, biodegradability is desired also for a grease composition used under a natural environment.

**[0003]** An aerogenerator is installed outside on the land or the sea and in case a grease composition used for an aerogenerator is leaked and released to a natural environment, a water quality or soil may be contaminated. Therefore biodegradability is desired also for a grease composition for an aerogenerator.

**[0004]** General aerogenerators consist of a blade (vane), tower (supporting column) and nacelle (body for generating electric power) and a grease composition is used for a spindle supporting bearing which rotates a blade by being subjected to wind power, a blade bearing used in a pitch revolving seat of the blade, a yaw rotation bearing used in a yaw rotation seat of the nacelle and the like.

**[0005]** The spindle supporting bearing, blade bearing and yaw rotation bearing as noted above are constantly subjected to micro-oscillation due to a change in a wind orientation or strength and a control of the blade or nacelle, and are in an environment in which abrasion or corrosion (fretting) easily arises. Therefore excellent fretting resistance is required for a grease composition for an aerogenerator.

**[0006]** In JP 2011-084646 A, there is disclosed a grease composition for bearings used in aerogenerators, comprising a base oil which has a kinematic viscosity of 10 to 70 mm<sup>2</sup>/s at 40°C and a pour point of -40°C or less, and a diurea compound as a thickener. However, biodegradability, fretting resistance and the like of a grease composition are not considered.

[0007] In JP 2008-208240 A, there is disclosed a biodegradable grease composition which can obtain biodegradability and extreme pressure property at low temperature by using a base oil comprising not less than 70% by mass of at least one selected from a polyol ester and a complex ester based on the whole amount of the base oil and having a kinematic viscosity at 40°C of 1 to 200 mm²/s. However, fretting resistance, the use in an aerogenerator and the like are not considered.

### SUMMARY OF THE INVENTION

**[0008]** An object of the present invention is to provide a biodegradable grease composition which is excellent in fretting resistance, extreme pressure property, low temperature property, and biodegradability as well, and has a small effect to the environment even if released to a natural environment.

**[0009]** The present invention relates to a biodegradable grease composition for an aerogenerator comprising a base oil composed of an ester oil having a kinematic viscosity at 40°C of 60 to 160 mm²/s and a thickener composed of a diurea compound obtained by allowing an amine mixture comprising a 4-8C alicyclic monoamine and a 20-24C aliphatic monoamine to react with a diisocyanate compound, wherein a content molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture is 7:3 to 9:1, the content of the thickener in a total amount of the base oil and the thickener is 7 to 11% by mass and a penetration of the biodegradable grease composition is 265 to 340.

[0010] It is preferable that a molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture is 8:2 to 9:1.

**[0011]** It is preferable that the alicyclic monoamine has 6 carbon atoms and the aliphatic monoamine has 22 carbon atoms.

**[0012]** It is further preferable that the biodegradable grease composition for an aerogenerator comprises a phosphorus-based antiwear agent.

**[0013]** According to the present invention, it is possible to provide a biodegradable grease composition for an aerogenerator which is excellent in fretting resistance, extreme pressure property, low temperature property, and biodegradability as well, and has a small effect to the environment even if released to a natural environment, by using a biodegradable grease composition comprising a base oil composed of an ester oil having a kinematic viscosity at 40°C within a predetermined range and a thickener composed of a predetermined diurea compound.

#### **DETAILED DESCRIPTION**

**[0014]** The biodegradable grease composition of the present invention is a biodegradable grease composition. Biodegradability refers to a nature of dissolving an organic substance by bacteria into carbon dioxide and water and into an inorganic compound, and ones having this nature are expressed as having biodegradability. Ease of microbial treatment is an index of biodegradability and as for a biodegradable grease composition, generally, a grease composition indicating a biodegradation degree of not less than 60% in a biodegradation degree test in accordance with an OECD method is regarded as a biodegradable grease composition and the same also applies herein.

#### 10 Base oil

15

20

25

30

40

45

50

55

**[0015]** Biodegradability of a grease composition largely depends on a base oil that is a main component of the biodegradable grease composition. In the present invention, for a biodegradable base oil, an ester oil chemically synthesized using natural fat, which can achieve both biodegradability and performance as a grease composition, as a raw material is used.

**[0016]** The ester oil is not limited particularly as long as it has biodegradability and has a kinematic viscosity at 40°C of 60 to 160 mm<sup>2</sup>/s and examples thereof include a fat acid ester, and a polyol ester, pentaerythritol tetraester and diester of a fatty acid and the like, and among these, a fatty acid ester is particularly preferable since biodegradability thereof is satisfactory.

**[0017]** A kinematic viscosity at 40°C of the base oil according to the present invention is not less than 60 mm²/s, preferably not less than 71 mm²/s, further preferably not less than 100 mm²/s. If the kinematic viscosity is less than 60 mm²/s, there is a tendency that extreme pressure property is deteriorated and an oil film becomes thinner. On the other hand, the kinematic viscosity at 40°C of the base oil is not more than 160 mm²/s, preferably not more than 150 mm²/s, further preferably not more than 120 mm²/s. If the kinematic viscosity exceeds 160 mm²/s, there is a tendency that fretting resistance and flowability are deteriorated.

**[0018]** The content of the base oil is preferably not less than 89% by mass, more preferably not less than 90% by mass based on the total content of the base oil and the thickener. If the content of the base oil is less than 89% by mass, there is a tendency that the grease composition becomes difficult to be biodegradable and low temperature property is deteriorated. On the other hand, the content of the base oil is preferably not more than 93% by mass, more preferably not more than 91 % by mass based on the total content of the base oil and the thickener. If the content of the base oil exceeds 93% by mass, there is a tendency that the biodegradable grease composition is softened and leaked.

#### Thickener

[0019] The thickener according to the present invention is composed of a diurea compound obtained by allowing an amine mixture comprising a 4-8C alicyclic monoamine and a 20-24C aliphatic monoamine to react with a diisocyanate compound. By using a diurea compound as a thickener, there is a tendency that an oil film becomes thicker and fretting resistance and heat resistance are improved.

**[0020]** The number of carbon atoms of the alicyclic monoamine is 4 to 8, preferably 5 to 7 and a cyclohexylamine having 6 carbon atoms is further preferable in view of its ease of availability. Examples of an alicyclic monoamine include cyclohexylamine, alkylcyclohexylamine and the like. Among these, cyclohexylamine is preferable since it is excellent in availability and heat resistance.

**[0021]** The number of carbon atoms of the aliphatic monoamine is 20 to 24, preferably 21 to 23, further preferably 22. If the number of carbon atoms of the aliphatic monoamine is less than 20, a thickening effect tends to decrease and an aliphatic monoamine having more than 24 carbon atoms is difficult to obtain.

**[0022]** A content molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture (alicyclic monoamine: aliphatic monoamine) is 7:3 to 9:1, more preferably 8:2 to 9:1, most preferably 8:2. The content molar ratio of the alicyclic monoamine and the aliphatic monoamine within this range enables a grease composition for an aerogenerator which is excellent in fretting resistance, extreme pressure property and low temperature property.

**[0023]** Examples of the diisocyanate compound include 4,4'-diphenylmethane-diisocyanate, 2,4-trilenediisocyanate, 2,6-trilenediisocyanate and the like. Among these, 4,4'-diphenylmethane-diisocyanate is preferable for its easy availability.

**[0024]** The reaction of the amine mixture with the diisocyanate compound can be carried out by various methods under various conditions, and it is preferable to carry out the reaction in the base oil since a diurea compound having highly uniform dispersibility can be obtained as the thickener. For example, the reaction may be carried out by adding the base oil containing the diisocyanate compound dissolved therein to the base oil in which the amine mixture has been dissolved, or by adding the base oil, in which the amine mixture has been dissolved, to the base oil containing the diisocyanate compound dissolved therein.

[0025] The reaction temperature and time in the above-mentioned reaction are not limited particularly, and may be the same as those used in usual similar reactions. The reaction temperature is preferably from 80°C to 100°C from the viewpoint of solubility and volatility of the amine mixture and diisocyanate. The reaction time is preferably less than 0.5 hour in view of improvement of production efficiency by shortening of the production period of time and also from the viewpoint of completing the reaction of the amine mixture and diisocyanate, and alternatively, the reaction may be conducted while mixing and elevating the temperature without determining the reaction time. The reaction of an amino group of the amine mixture and an isocyanate group of the diisocyanate compound proceeds quantitatively, and a preferred ratio thereof is 1 mole of the diisocyanate compound to 2 mole of the amine mixture.

[0026] The diurea compound which is a reaction product obtained by the above-mentioned reaction is a mixture of a diurea compound comprising any of (a) a diurea compound in which both isocyanate groups of a diisocyanate compound are reacted with an alicyclic amine in the amine mixture; (b) a diurea compound in which both isocyanate groups of a diisocyanate compound are reacted with an aliphatic amine in the amine mixture; and (c) a diurea compound in which one of isocyanate groups of a diisocyanate compound is reacted with an alicyclic amine and the other is reacted with an aliphatic amine. It is noted that a diurea compound used in the present invention further include a diurea compound which is a reaction product obtained by synthesizing each of the above-mentioned diurea compounds (a) to (c) and mixing these compounds.

[0027] The content of the above thickener is not less than 7% by mass, preferably not less than 9% by mass based on the total amount of the base oil and the thickener. If the content of the thickener is less than 7% by mass, there is a tendency that the biodegradable grease composition is softened and leaked. On the other hand, the content of the thickener is not more than 11% by mass, preferably not more than 10% by mass based on the total amount of the base oil and the thickener. If the content of the thickener exceeds 11% by mass, a biodegradation rate of the biodegradable grease composition tends to decrease.

#### Additives

10

15

20

25

30

35

40

45

50

55

**[0028]** The biodegradable grease composition of the present invention may comprise various additives such as an antioxidant, an extreme pressure agent, an antiwear agent, a dye, a color stabilizer, a viscosity improver, a structure stabilizer, a metal deactivator, a viscosity index improver, a dispersing agent and a rust-preventing agent in proper amounts to such an extent not to impair the effect of the present invention. It is noted that considering an effect to the environment, it is preferable that additives comprising heavy metal are not contained. When these additives are contained in the biodegradable grease composition, the amount thereof in the biodegradable grease composition is preferably 0.5 to 10 parts by mass based on the total of 100 parts by mass of the base oil and the thickener.

**[0029]** Examples of the antiwear agent include methylenebis (dithiocarbamate), a sulfur-based antiwear agent, a phosphorus-based antiwear agent and the like. Among these, it is more preferable to use a phosphorus-based antiwear agent since it is excellent in antiwear property.

[0030] Specific examples of the phosphorus-based antiwear agent include zinc dialkyl dithiophosphate; phosphites represented by tributyl phosphite, trioleilphosphite and the like; phosphates represented by tricresylphosphate, dilauryl acid phosphate and the like; amine phosphates represented by phosphoric acid dibutyloctyl amine salt, phosphoric acid dilauryloctyl amine salt and the like; phosphorothionates represented by triphenyl phosphorothionate, alkylated phosphorothionate and the like; solid lubricants represented by calcium phosphate; and diphenyl hydrogen phosphites. In the present invention, commercial available phosphorus-based antiwear agents can be also used. Among these, amine phosphates are preferable since the burden on the environment is small and a specific example thereof includes Lubrizol 4320 FG manufactured by The Lubrizol Corporation and the like.

**[0031]** When the biodegradable grease composition comprises an antiwear agent, the content thereof based on the total of 100 parts by mass of the base oil and the thickener is preferably not less than 0.1 part by mass, more preferably 0.5 to 5 parts by mass, further preferably 1 to 3 parts by mass. If the content of the antiwear agent is less than 0.1 part by mass, an effect obtained by using the antiwear agent tends not to be obtained. On the other hand, if the content of the antiwear agent exceeds 5 parts by mass, biodegradability tends to be deteriorated.

**[0032]** Examples of the extreme pressure agent include a sulfur-based extreme pressure agent, a phosphorus-based extreme pressure agent and the like. Among these, it is preferable that the biodegradable grease composition comprises a sulfur-based extreme pressure agent since it can impart an extreme pressure effect in a small amount.

**[0033]** When the biodegradable grease composition comprises an extreme pressure agent, the content thereof based on the total of 100 parts by mass of the base oil and the thickener is preferably 0.1 to 3 parts by mass, more preferably 0.5 to 2 parts by mass. If the content of the extreme pressure agent is less than 0.1 part by mass, an effect obtained by using the extreme pressure agent tends not to be obtained. On the other hand, if the content of the extreme pressure agent exceeds 3 parts by mass, a raw material cost tends to be high.

**[0034]** The worked penetration of the biodegradable grease composition of the present invention is 265 to 340, preferably 270 to 320, more preferably 280 to 315. If the worked penetration exceeds 340, the biodegradable grease

composition tends to be easily leaked from the inside of a bearing. On the other hand, if the worked penetration is less than 265, there is a tendency that a torque of the grease-applied parts increases and a service life is decreased because of seizure by lowering of flowability.

**[0035]** While the biodegradable grease composition for an aerogenerator of the present invention can be used for a spindle supporting bearing, a blade bearing, a yaw rotation bearing and the like of an aerogenerator, it is preferable to use the biodegradable grease composition of the present invention for a blade bearing of an aerogenerator since it has a low viscosity and is excellent in preventing fretting due to micro-oscillation.

#### **EXAMPLE**

10

20

25

[0036] In the following, while the present invention will be explained in more detail by use of Examples, the present invention is not limited thereto.

[0037] The following raw materials were used in Examples.

15 Base oil

#### [0038]

Ester oil 1: Synative ES 3345 (fatty acid ester, kinematic viscosity (40°C): 112 mm<sup>2</sup>/s) manufactured by BASF Japan Ltd.

Ester oil 2: Priolube 2089 (fatty acid ester, kinematic viscosity (40°C): 46 mm<sup>2</sup>/s) manufactured by Croda Japan KK Ester oil 3: Synative ES 3157 (fatty acid ester, kinematic viscosity (40°C): 46 mm<sup>2</sup>/s) manufactured by BASF Japan Ltd.

Ester oil 4: Synative ES 1200 (fatty acid ester, kinematic viscosity (40°C): 1200 mm²/s) manufactured by BASF Japan Ltd.

Ester oil 5: Synative ES TMP 05/320 (fatty acid ester, kinematic viscosity (40°C): 326 mm<sup>2</sup>/s) manufactured by BASF Japan Ltd.

Thickener

30

35

40

Diisocyanate compound

**[0039]** MDI: Millionate MT-F (4,4'- diphenyl methane diisocyanate, molecular weight: 250.25) manufactured by Nippon Polyurethane Industry Co., Ltd.

Amine

[0040] Behenylamine: Amine VB-S (22C aliphatic amine, molecular weight: 325.62) manufactured by NOF CORPORATION

[0041] Stearylamine: Amine HT flake (18C aliphatic amine, molecular weight: 269.51) manufactured by LION SPE-CIALITY CHEMICALS CO., LTD. Cyclohexylamine: CHA (6C alicyclic amine, molecular weight: 99.17) manufactured by New Japan Chemical Co., Ltd.

Additives

45

50

[0042] Extreme pressure agent: Addition RC8400 (sulfur-based white color solid lubricant) manufactured by Rhein Chemie Rheinau GmbH

[0043] Ca sulphonate: NA-SUL CA-770FG manufactured by King Industries Inc.

**[0044]** Rust-preventing agent 1: Alcatase T (oxazoline-based rust-preventing agent) manufactured by The Dow Chemical Company

[0045] Rust-preventing agent 2: Nonion OP-80R (sorbitan monooleate) manufactured by NOF CORPORATION

Antiwear agent: Lubirizol 4320FG (amine phosphate) manufactured by The Lubrizol Corporation

55 Examples and Comparative Examples

**[0046]** According to the formulation shown in Tables 1 and 2, grease compositions were respectively prepared. Firstly, a part of calcium sulphonate (10% by mass based on the thickener) and each of amines were added to a base oil, the

mixture was maintained at 80 to 90°C, a diisocyanate compound was further added thereto, the mixture was heated to 160°C while stirring and an extreme pressure agent was further added thereto. The mixture was cooled while stirring and homogenized via a homogenizer treatment (pressure: about 300 bar) to prepare a base grease. Then, the remaining calcium sulphonate and other additives were added thereto and the mixture was stirred and defoamed to prepare respective test grease compositions. The obtained test grease compositions were subjected to the following evaluations. The results are shown in Tables 1 and 2.

<Measurement of worked penetration>

[0047] The worked penetration is a value obtained by dropping a cone mounted on a cone penetration meter into the test grease compositions under environment of 25°C, measuring a depth (mm) of 5-second invasion of the cone into the grease, and then multiplying the measured depth by 10 in accordance with JIS K2220-7.

<Measurement of extreme pressure>

**[0048]** The weld load of the test grease compositions was measured with the method of ASTM D2596 (high-speed four ball test) under the following test conditions. The larger the value of weld load is, the more excellent the extreme pressure property is. It is noted that the performance target value is 2452 N or more.

Number of revolutions: 1770 rpm

Test temperature: room temperature (25°C)

Test time: 10 seconds

15

25

30

35

45

50

55

[0049] <Fafnir wear volume>

**[0050]** The fretting resistance test was conducted in accordance with ASTM D4170 and a Fafnir wear volume (mg) was measured from a mass difference between before and after the test. The less the Fafnir wear volume is, the more excellent the fretting resistance is. It is noted that the performance target value is 1.0 mg or less.

<Low temperature property test>

**[0051]** By use of a rheometer device (ARES-RDA3 manufactured by TA Instruments Japan Inc.), a torque at starting and a torque while rotating were measured under the condition where a shear rate becomes  $10s^{-1}$  after setting a gap (0.5 mm) between a rotating upper plate and a fixed lower plate, sandwiching each grease composition between the gap, and maintaining an environment of -20°C. The less the both torque are, the more excellent the low temperature property is. It is noted that the performance target value of the torque at starting is 15 mN·m or less and the performance target value of the torque while rotating is 5 mN·m or less.

40 <Shear stability test>

**[0052]** The worked penetration of each grease composition was measured after applying shearing force for two hours in accordance with ASTM D1831. The smaller the value of the worked penetration is, the more excellent the shear stability is. It is noted that the performance target value is 375 or less.

<Biodegradability test>

**[0053]** The biodegradation rate (%) of the test grease composition of Example 1 was measured in accordance with OECD 301C. Based on the biodegradation rate of Example 1, biodegradability of other Examples and Comparative Examples was calculated in the following formula. The biodegradation rate of 60% or more is represented as  $\bigcirc$  and the biodegradation rate of less than 60% is represented as  $\times$ .

Biodegradability (%) = (biodegradation rate of Example 1)  $\times$  (content of base oil of each grease composition) / (content of base oil of Example 1)

# <Film thickness measuring test>

**[0054]** Oil film forming property of each grease composition was evaluated at room temperature using an oil film thickness measuring device to which optical interferometry is applied and which is manufactured by PCS Instruments. A 3/4 inch diameter steel ball of the bearing was set at a load of 20 N on a surface of a hard glass having a diameter of about 10 cm on which each grease composition was applied in a film thickness of 1 mm, and the hard glass was rotated so that the rolling speed of the contacting raceway portion became 1.00 m/s. Then, the rolling speed was gradually decreased to 0.10 m/s in 60 seconds and the oil film thickness at which the rolling speed became 0.10 m/s was regarded as the ELH oil film thickness of each grease composition. It is noted that the performance target value is 150 nm or more.

10

5

	[Table 1]					
	Examples					
	1	2	3	4	5	6
Compounding amount (part by mass)						
Base oil						
Ester oil 1	90.92	59.03	77.56	71.48	58.44	83.0
Ester oil 2						
Ester oil 3		31.89		18.43	31.47	
Ester oil 4			13.36			6.83
Ester oil 5						
(Viscosity of base oil (40°C))	(112)	(78)	(154)	(92)	(78)	(130
Thickener						
MDI	4.20	4.20	4.20	5.12	4.32	5.1
Cyclohexylamine	2.68	2.68	2.68	3.64	2.40	3.6
Behenylamine	2.20	2.20	2.20	1.33	3.37	1.3
Stearylamine						
(Molar ratio of alicyclic amine : aliphatic amine)	(8:2)	(8:2)	(8:2)	(9:1)	(7:3)	(9:1
(Total amount of thickener)	(9.08)	(9.08)	(9.08)	(10.09)	(10.09)	(10.0
(Base oil + Thickener)	(100)	(100)	(100)	(100)	(100)	(100
Additives						
Extreme pressure agent	1.01	1.01	1.01	1.01	1.01	1.0
Ca sulphonate	4.94	4.94	4.94	4.94	4.94	4.9
Rust-preventing agent 1	2.02	2.02	2.02	2.02	2.02	2.0
Rust-preventing agent 2	1.01	1.01	1.01	1.01	1.01	1.0
Antiwear agent	1.01	1.01	1.01	1.01	1.01	1.0
Evaluation result						
Worked penetration	311	280	296	305	297	30
Extreme pressure property (N)	2452	2452	2452	2452	2452	245
Fafnir wear volume (mg)	0.3	0.4	0.9	8.0	0.7	0.6
Low temperature test (mN·m)						
Torque at starting	10.8	10.9	13.5	11.5	12.2	13.
Torque while rotating	3.5	3.6	4.6	3.2	3.7	4.
Shear stability	369	356	353	371	368	35
Biodegradability (%)	0	$\circ$	$\circ$	$\circ$	0	0
EHL oil film thickness (nm)	227	172	281	233	238	25

[Table 2]

	Comparative Examples					
	1	2	3	4	5	6
Compounding amount (part by mass)						
Base oil						
Ester oil 1	-	68.18	-	87.90	88.90	84.86
Ester oil 2	90.92	-	-	-	-	-
Ester oil 3	-	-	-	-	-	-
Ester oil 4	-	22.73	-	-	-	-
Ester oil 5	-	-	90.92	-	-	-
(Viscosity of base oil (40°C))	(46)	(197)	(326)	(112)	(112)	(112)
Thickener						
MDI	4.20	4.20	4.20	5.60	5.38	6.12
Cyclohexylamine	2.68	2.68	2.68	3.57	3.41	2.43
Behenylamine	2.20	2.20	2.20	2.93	-	-
Stearylamine	-	-	-	-	2.31	6.59
(Molar ratio of alicyclic amine: aliphatic amine)	(8:2)	(8:2)	(8:2)	(8:2)	(8:2)	(5:5)
(Total amount of thickener)	(9.08)	(9.08)	(9.08)	(12.1)	(11.1)	(15.14)
(Base oil + Thickener)	(100)	(100)	(100)	(100)	(100)	(100)
Additives						
Extreme pressure agent	1.01	1.01	1.01	1.01	1.01	1.01
Ca sulphonate	4.94	4.94	4.94	4.94	4.94	4.94
Rust-preventing agent 1	2.02	2.02	2.02	2.02	2.02	2.02
Rust-preventing agent 2	1.01	1.01	1.01	1.01	1.01	1.01
Antiwear agent	1.01	1.01	1.01	1.01	1.01	1.01
Evaluation result						
Worked penetration	311	295	274	300	279	310
Extreme pressure property (N)	1961	2452	2452	2452	2452	2452
Fafnir wear volume (mg)	0.6	1.5	17.5	26.3	1.7	35.7
Low temperature test (mN·m)						
Torque at starting	8.7	16.8	25.3	19.0	20.9	18.5
Torque while rotating	2.1	6.0	6.6	4.0	3.9	3.7
Shear stability	381	346	352	344	345	357
Biodegradability (%)	0	$\circ$	$\circ$	$\circ$	0	0
EHL oil film thickness (nm)	88	320	400	286	267	316

**[0055]** From the results shown in Tables 1 and 2, it is found that a grease composition comprising a base oil composed of an ester oil having a kinematic viscosity at 40°C in a predetermined range and a thickener composed of a predetermined diurea compound is a biodegradable grease composition for an aerogenerator which is excellent in fretting resistance, extreme pressure property, low temperature property, and biodegradability as well, and has a small effect to the environment even if released to a natural environment.

# 50 Claims

45

- A biodegradable grease composition for an aerogenerator comprising a base oil composed of an ester oil having a
  kinematic viscosity at 40°C of 60 to 160 mm<sup>2</sup>/s and a thickener composed of a diurea compound obtained by allowing
  an amine mixture comprising a 4-8C alicyclic monoamine and a 20-24C aliphatic monoamine to react with a diisocyanate compound,
  - wherein a content molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture is 7:3 to 9:1,

the content of the thickener in a total amount of the base oil and the thickener is 7 to 11% by mass and a penetration of the biodegradable grease composition is 265 to 340.

- 2. The biodegradable grease composition for an aerogenerator of claim 1, wherein a molar ratio of the alicyclic monoamine and the aliphatic monoamine in the amine mixture is 8:2 to 9:1.
  - **3.** The biodegradable grease composition for an aerogenerator of claim 1 or 2, wherein the alicyclic monoamine has 6 carbon atoms and the aliphatic monoamine has 22 carbon atoms.
- **4.** The biodegradable grease composition for an aerogenerator of any of claims 1 to 3, wherein the biodegradable grease composition for an aerogenerator comprises a phosphorus-based antiwear agent.



# **EUROPEAN SEARCH REPORT**

Application Number

EP 15 19 2511

	des b	prevets			EP 15 19 2
_					1
<u> </u>	- 1	DOCUMENTS CONSID	ERED TO BE RELEVANT	1	
Ce	ategory	Citation of document with in of relevant passa	idication, where appropriate, ages	Relevant to claim	CLASSIFICATION OF 1 APPLICATION (IPC)
X		AL) 10 July 2014 (2 * paragraphs [0001]	, [0064], [0065], 0119] - [0122], [0117]	1,2,4	INV. C10M169/02
		Retrieved from the URL:https://www.ecl	<pre>lube.com/userfiles/prod %20Eng.%20Guide_ENG_fin 02-29]</pre>		
l <sub>x</sub>		US 2007/072777 A1 (	OKAMURA SEIJI [JP] ET	3	
		AL) 29 March 2007 (			TECHNICAL FIELDS SEARCHED (IPC)
A	ı	* claim 1 * * page 4 - page 5; * paragraph [0002]	example 3; table 1 *	1,2,4	C10M C10N
A	,	JP 2003 306687 A (N 31 October 2003 (20 * the whole documen * table 1 *	03-10-31)	1-4	
A	\   	US 2003/176298 A1 (AL) 18 September 20 * paragraphs [0002] [0104] * claims 1, 3, 9, 1	, [0131], [0103],	1-4	
			-/		
1		The present search report has b	peen drawn up for all claims		
1		Place of search	Date of completion of the search		Examiner
4C01)		Munich	14 March 2016 Ka		uza, Nora
32 (P0	CA	ATEGORY OF CITED DOCUMENTS	T : theory or principle		
EPO FORM 1503 03.82 (P04C01)	Y : parti docu A : tech O : non-	oularly relevant if taken alone cularly relevant if combined with anoth ment of the same category nological background written disclosure mediate document	E : earlier patent document, but published on, or after the filing date		



Category

# **EUROPEAN SEARCH REPORT**

Citation of document with indication, where appropriate, of relevant passages

Application Number

EP 15 19 2511

CLASSIFICATION OF THE APPLICATION (IPC)

Relevant to claim

10		
15		
20		
25		
30		
35		
40		
45		

50

55

	or relevant pass	ages	10	Oldilli			
A	EP 2 489 721 A1 (KY 22 August 2012 (201 * paragraphs [0002] [0048] * * page 4; compound * example 4; table	2-08-22) , [0025], [00	) 1-4	1			
A,P	EP 2 829 593 A1 (JT GREASE CO LTD [JP]) 28 January 2015 (20 * paragraphs [0012] [0004] * claim 1 * & JP 61 155496 A 15 * the whole documer & WO 2013/141365 A1 26 September 2013 (* the whole documer	015-01-28) , [0003], [00 July 1986 (198 st * . (JTEKT CORP) 2013-09-26)	920],	2,4			
A	US 2013/331306 A1 ( 12 December 2013 (2 * paragraphs [0001] [0043] *	013-12-12)			TECHNICAL FIELDS SEARCHED (IPC)		
А	US 2004/242439 A1 (AL) 2 December 2004 * sentences 0001, 6	(2004-12-02)		4			
Α	CN 1 940 039 A (MIN 4 April 2007 (2007- * the whole documer	04-04)	3				
	The present search report has	·					
		Date of completio	npletion of the search		Examiner Nova		
	Munich				a, Nora		
CATEGORY OF CITED DOCUMENTS  X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background		E: f her D: L:c	T: theory or principle underlying the invention E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons				
Munich  CATEGORY OF CITED DOCUMENTS  X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document				atent family, corr			

# ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.

EP 15 19 2511

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

14-03-2016

)	Patent document cited in search report	Publication date	Patent family member(s)	Publication date	
5	US 2014193110 A1	10-07-2014	CN 103814119 A DE 112012003999 T5 JP 2013082882 A US 2014193110 A1 WO 2013046598 A1	21-05-2014 21-08-2014 09-05-2013 10-07-2014 04-04-2013	
)	US 2007072777 A1	29-03-2007	JP 4968825 B2 JP 2007112998 A US 2007072777 A1	04-07-2012 10-05-2007 29-03-2007	
	JP 2003306687 A	31-10-2003	NONE		
	US 2003176298 A1	18-09-2003	NONE		
5	EP 2489721 A1	22-08-2012	CA 2777188 A1 CN 102549127 A EP 2489721 A1 JP 5664890 B2 JP 2011084646 A US 2012196781 A1 WO 2011046201 A1	21-04-2011 04-07-2012 22-08-2012 04-02-2015 28-04-2011 02-08-2012 21-04-2011	
5	EP 2829593 A1	28-01-2015	CN 104204169 A EP 2829593 A1 JP 5865146 B2 JP 2013194210 A US 2015078687 A1 WO 2013141365 A1	10-12-2014 28-01-2015 17-02-2016 30-09-2013 19-03-2015 26-09-2013	
)	US 2013331306 A1	12-12-2013	CN 103403137 A DE 112012001102 T5 JP 5727276 B2 JP 2012197401 A US 2013331306 A1 WO 2012121212 A1	20-11-2013 19-12-2013 03-06-2015 18-10-2012 12-12-2013 13-09-2012	
5	US 2004242439 A1	02-12-2004	JP 4334915 B2 JP 2004352858 A US 2004242439 A1	30-09-2009 16-12-2004 02-12-2004	
)	CN 1940039 A	04-04-2007	NONE		
OFM P0459					

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

#### REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

# Patent documents cited in the description

• JP 2011084646 A **[0006]** 

• JP 2008208240 A [0007]