# (11) EP 3 109 300 A1

(12)

# EUROPEAN PATENT APPLICATION

published in accordance with Art. 153(4) EPC

(43) Date of publication: 28.12.2016 Bulletin 2016/52

(21) Application number: 15751845.7

(22) Date of filing: 18.02.2015

(51) Int Cl.: C10M 169/04 (2006.01)

C10M 125/10 (2006.01) C10N 10/12 (2006.01) C10N 30/06 (2006.01) C10N 50/10 (2006.01) C10M 107/50 (2006.01) C10N 10/04 (2006.01) C10N 20/00 (2006.01) C10N 40/04 (2006.01)

(86) International application number:

PCT/JP2015/054383

(87) International publication number: WO 2015/125805 (27.08.2015 Gazette 2015/34)

(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

(30) Priority: 18.02.2014 JP 2014028681

(71) Applicant: Kyodo Yushi Co., Ltd. Kanagawa 251-8588 (JP)

(72) Inventors:

HIROOKA, Iwaki
 Fujisawa-shi, Kanagawa 251-8588 (JP)

YOSHINARI, Terasu
 Fujisawa-shi, Kanagawa 251-8588 (JP)

(74) Representative: Hart-Davis, Jason et al Cabinet Beau de Loménie 158, rue de l'Université 75340 Paris Cedex 07 (FR)

### (54) SILICONE GREASE COMPOSITION

(57) The invention provides a silicone grease composition where the high friction properties are improved, with no decrease of the wear preventive characteristics. The silicone grease composition contains a thickener, a

base oil containing a silicone oil in an amount of 50 mass% or more of the total mass of the base oil, and a metallic oxide with a Mohs hardness of 6 or less as a friction modifier.

EP 3 109 300 A1

#### Description

#### [Technical Field]

**[0001]** The present invention relates to a silicone composition. More particularly, the invention relates to a silicone grease composition suitably used for the parts to be lubricated, to be more specific, the lubrication parts of the clutch, the torque limiter mechanism and the like where high coefficient of friction and excellent wear preventive characteristics are needed.

### [Background Art]

15

20

30

35

45

50

55

**[0002]** In consideration of the global environmental problems, weight reduction of the automobile has been advancing. In line with this tendency, a variety of portions tend to use more clutches and torque limiter mechanisms than ever. Of such a variety of clutches and torque limiter mechanisms using the grease composition in the automobile, the engine starter clutch is exposed to the highest torque and the severest operating conditions.

**[0003]** The clutch of the engine starter is composed of a clutch outer, a clutch inner, a roller disposed in a wedge-like space formed between the clutch outer and the clutch inner to transmit the rotation of the clutch outer to the clutch inner, and a spring which works to urge the roller toward the narrower side of the wedge-like space. When the clutch outer is rotated, the roller moves toward the narrower side of the wedge-like space and is then caught between the clutch outer and the clutch inner, thereby transmitting the rotation to the clutch inner (torque-transmitted state). Accordingly, the grease composition used for such portions requires a high coefficient of friction to prevent the slippage of the clutch outer, the clutch inner and the roller.

[0004] The following silicone grease compositions are conventionally known: a grease for the overrunning clutch (JP (Hei) 7-35824 B) where a base oil comprising a silicone oil has a coefficient of friction of 0.18 or more; grease compositions (JP (Hei) 5-230486 A and JP (Hei) 6-279777 A) comprising as the base oil organopolysiloxanes having a predetermined ratio of phenyl group or methyl group; and a traction grease composition (JP 2003-176489 A) comprising finely-divided particles of metallic oxide, a thickener, and a base oil containing a dimethyl silicone oil with a predetermined kinematic viscosity in an amount of 1 to 40 mass%. Those grease compositions make use of the advantages of the silicone oils that the coefficients of friction are high, and in addition, the surface tensions of the silicone oils range from 20 to 25 dyn/cm2, which are lower than those of other oils, so that a lubrication film may not readily be formed, thereby easily attaining to boundary lubrication.

**[0005]** When the rotational speed of the clutch inner becomes higher than that of the clutch outer, the roller automatically moves toward a wider side of the wedge-like space as compressing the spring, thereby stopping the transmission of the rotation (torque non-transmitted state). Under such conditions, the clutch outer, the clutch inner and the roller reach a state of slippage due to generation of the relative rotation. In light of this, the grease composition used for such portions is required to have wear preventive characteristics.

### [Summary of Invention]

### 40 [Technical Problem]

[0006] As previously explained, the clutch and the torque limiter mechanism are required to be smaller in size and lighter in weight in line with the recent tendency toward weight reduction in the automobile, and on the other hand, they are exposed to severer operating conditions. Especially, satisfactory torque transmission properties are demanded even when the operating conditions become more and more tough. For that reason, the grease composition is required to have a higher coefficient of friction. At the same time, the wear preventive characteristics are also needed when the grease composition is used for the clutch and the torque limiter mechanism.

**[0007]** Accordingly, an object of the invention is to provide a silicone grease composition having improved frictional characteristics, without deterioration of the wear preventive characteristics.

#### [Solution to Problem]

**[0008]** To solve the above-mentioned problems, the inventors of the present invention contrived a grease composition having an increased coefficient of friction without decrease of the wear preventive characteristics by adding as the friction modifier a metallic oxide with a Mohs hardness of 6 or less.

**[0009]** Namely, the invention provides a grease composition as shown below.

1. A silicone grease composition comprising a thickener, a base oil comprising a silicone oil in an amount of 50

mass% or more of the total mass of the base oil, and a friction modifier comprising a metallic oxide with a Mohs hardness of 6 or less

- 2. The silicone grease composition described in the above-mentioned item 1, wherein the metallic oxide has a Mohs hardness of 2 to 6.
- 3. The silicone grease composition described in the above-mentioned item 1 or 2, wherein the metallic oxide is magnesium oxide, zinc oxide or molybdenum oxide.
- 4. The silicone grease composition described in any one of the above-mentioned items 1 to 3, wherein the metallic oxide is contained in an amount of 0.1 to 10 mass% based on the total mass of the composition.

#### [Effects of Invention]

[0010] According to the invention, a silicone grease composition where the frictional characteristics are improved without decrease of the wear preventive characteristics can be provided.

### [Description of Embodiments]

20 <Base oil>

5

10

30

35

40

45

50

[0011] A silicone oil is used for the base oil in the grease composition according to the invention.

[0012] Specific examples of the silicone oil include dimethyl silicone oil, methylphenyl silicone oil (phenyl-modified silicone oil), methyl hydrogen silicone oil, polyether-modified silicone oil, aralkyl-modified silicone oil, fluoroalkyl-modified silicone oil, alkyl-modified silicone oil, fatty acid ester-modified silicone oil, and the like. Of the above silicone oils, dimethyl silicone oil and methylphenyl silicone oil are preferably used. The silicone oil represented by the following general formula (1) is particularly preferable:

$$(CH_3)_3SiO-[-Si(R1)(R2)-O-]n - Si(CH_3)_3$$
 (1)

wherein R1 and R2 are each independently methyl group or phenyl group, the ratio of methyl group to the whole organic groups being 50 to 100 mol.%.

[0013] The kinematic viscosity of the silicone oil is not particularly limited, but may preferably be in the range of 20 to 10,000 mm²/s, more preferably 50 to 2,000 mm²/s at 25°C. When the kinematic viscosity of the silicone oil is less than 20 mm²/s, the base oil may easily separate from the grease. When the kinematic viscosity exceeds 10,000 mm²/s, the torque will increase at low temperatures due to the viscosity resistance. In any case, the results are practically undesirable. [0014] In the above-mentioned formula (1), R1 and R2 are each independently methyl group or phenyl group, the ratio of the methyl group to the whole organic groups being 50 to 100 mol.%. When the molar ratio of the methyl group to the whole organic groups is less than 50 mol.%, the viscosity greatly varies depending on the temperature, and the pour point increases and the torque becomes large at low temperatures. The ratio of the methyl group to the whole organic groups may preferably be 60 to 100 mol.%, more preferably 80 to 98 mol.%, and still more preferably 90 to 95 mol.%. [0015] The silicone oil may preferably be contained in an amount of 55 to 90 mass%, and more preferably 70 to 90 mass%, with respect to the total mass of the composition. Such a content of the silicone oil can favorably lead to high coefficient of friction.

[0016] The silicone oil may be used in combination with other base oil components so long as the performance of the silicone oil may not be impaired. One or more base oil components selected from the group consisting of mineral oils, poly  $\alpha$ -olefins, polybutene, alkylbenzene, animal and vegetable oils, organic acid esters, diesters, polyol esters, polyalkylene glycols, polyvinyl ethers, polyphenyl ethers, and alkylphenyl ethers can be used. The amount of other base oil component(s) that can be used in combination with the silicone oil may preferably be 0 to 50 mass% with respect to the mass of the silicone oil used for the base oil. In order not to decrease the high coefficient of friction, the amount of other base oil component(s) may preferably be 0 to 20 mass%, more preferably 0 to 10 mass%, with respect to the mass of the lubricating base oil used in the invention. It is most preferable to use no other base oil component.

### <Thickener>

55

**[0017]** The thickener used for the grease composition of the invention is not particularly limited. Specific examples include soap type thickeners such as lithium soaps and lithium complex soaps, urea type thickeners such as diurea compounds, inorganic thickeners such as organoclay and silica, organic thickeners such as polytetrafluoroethylene and

melamine cyanurate, and the like. At least one selected from the above-mentioned group may be used. It is preferable to select from the group consisting of silica, lithium soaps, lithium complex soaps and urea compounds. In particular, the urea compounds are preferable. As the urea type thickener, diurea compounds are preferable, and in particular the diurea compounds represented by the following general formula (2) are preferred:

### R3-NHCONH-R4-NHCONH-R5 (2)

wherein R3 and R5, which may be the same or different from each other, each represent a residue of monovalent hydrocarbon groups having 4 to 20 carbon atoms, for example, aliphatic hydrocarbon groups, alicyclic hydrocarbon groups and aromatic hydrocarbon groups; and R4 is a bivalent aromatic hydrocarbon group having 6 to 15 carbon atoms. [0018] The above-mentioned diurea thickener can be obtained by reacting a predetermined diisocyanate with a predetermined monoamine in the base oil, for example. Specific examples of the preferable diisocyanate include diphenylmethane-4,4'-diisocyanate and tolylene diisocyanate. Examples of the monoamine include aliphatic amines, aromatic amines and alicyclic amines, or the mixtures thereof. Specific examples of the aliphatic amines include octylamine, dodecylamine, hexadecylamine, octadecylamine, octadecylamine. Specific examples of the aromatic amines include aniline and p-toluidine. Specific examples of the alicyclic amines include cyclohexylamine. Of the above-mentioned monoamines, cyclohexylamine, octylamine, dodecylamine, hexadecylamine, octadecylamine, or the mixture thereof may preferably be used for preparation of the diurea thickener. In particular, alicyclic aliphatic diurea compounds are preferred which can be obtained by using cyclohexylamine together with octylamine, dodecylamine, hexadecylamine, octadecylamine or the mixture thereof. Further, alicyclic aliphatic diurea compounds obtained from cyclohexylamine and octadecylamine, or the mixtures thereof are particularly preferable. A mixture of the following three kinds of diurea compounds represented by formula (2-1), (2-2) and (2-3) (where R is octadecyl group) is most preferable.

### [Chemical 1]

$$\sim$$
 NHCONH  $\sim$  CH<sub>2</sub>  $\sim$  NHCONH  $\sim$  (2-1)

$$-$$
NHCONH $-$ CH $_2$ -NHCONH $-$ R (2-2)

$$R-NHCONH-CH_2-NHCONH-R$$
 (2-3)

**[0019]** The content of the thickener, which may appropriately be adjusted in accordance with the desired consistency is generally 2 to 35 mass%, preferably 5 to 30 mass%, and more preferably 10 to 25 mass%.

**[0020]** When the content of the thickener is less than 2 mass%, the resultant product becomes a liquid, which cannot be used as a grease. When the content of the thickener exceeds 35 mass%, the resultant grease is not practically desirable because the grease is so hard that the torque will increase at low temperatures.

#### <Friction modifier>

5

10

15

20

25

30

35

40

45

50

55

**[0021]** The grease composition of the invention comprises as the friction modifier a metallic oxide with a Mohs hardness of 6 or less.

**[0022]** The addition of the metallic oxide as the friction modifier can increase the coefficient of friction because the oil film becomes easy to break due to the metallic oxide entering into the surfaces to be lubricated. Even when the lubrication member is made of steel, use of the metallic oxide with a Mohs hardness of 6 or less can increase the coefficient of friction, with damage to the steel being minimized.

[0023] More specifically, magnesium oxide, potassium oxide, calcium oxide, scandium oxide, titanium oxide, vanadium oxide, chromium oxide, manganese oxide, iron oxide, cobalt oxide, nickel oxide, copper oxide, zinc oxide, gallium oxide, germanium oxide, strontium oxide, yttrium oxide, zirconium oxide, niobium oxide, molybdenum oxide, technetium oxide, ruthenium oxide, rhodium oxide, palladium oxide, silver oxide, indium oxide, tin oxide, antimony oxide, tellurium oxide, barium oxide, hafnium oxide, tantalum oxide, tungsten oxide, rhenium oxide, lead oxide, and silicon oxide can be used. Particularly, magnesium oxide, zinc oxide and molybdenum oxide are preferable. The Mohs hardness of the metallic

oxide is preferably 6 or less, more preferably 2 to 6, and still more preferably 4 to 6. The Mohs hardness of more than 6 is not favorable because the abrasion will become harsh during the idling operation in the torque non-transmitted state. **[0024]** The metallic oxide may preferably have an average particle diameter of 10  $\mu$ m or less, more preferably 5  $\mu$ m or less, and still more preferably 2  $\mu$ m or less. When the average particle diameter exceeds 10  $\mu$ m, it becomes difficult for the metallic oxide particles to come between the surfaces to be lubricated, and therefore a sufficient effect cannot be expected. The BET conversion method by N<sub>2</sub>-adsorption may be used to determine the average particle diameter herein used.

**[0025]** The metallic oxide may preferably be contained in an amount of 0.1 to 10.0 mass%, more preferably 0.3 to 7.0 mass%, still more preferably 0.5 to 5.0 mass%, and most preferably 1.0 to 3.0 mass%, based on the total mass of the composition. When the content of the metallic oxide is less than 0.1 mass%, a sufficient effect cannot be expected. When the content of the metallic oxide exceeds 10.0 mass%, the cost will be disadvantageous because the effect is saturated.

<Other additives>

10

15

20

25

30

35

40

45

55

**[0026]** Other additives conventionally used for grease, such as an antioxidant, rust inhibitor, metal deactivator, detergent dispersant, extreme pressure agent, antifoaming agent, demulsifier, oilness improver, solid lubricant and the like may be incorporated. Those auxiliary additives may be used alone or in combination. The auxiliary additives may be added if necessary, and in this case, the contents may be 0.01 to 10 mass% generally. However, the contents of those additives are not particularly limited as far as the effects of the invention are not degraded.

<Consistency>

[0027] The grease composition of the invention may preferably have a worked penetration of 200 to 400, more preferably 230 to 380, and most preferably 250 to 350.

**[0028]** According to one preferable embodiment of the invention, the silicone grease composition comprises; a mixture of three kinds of diurea compounds represented by the above-mentioned formulas (2-1), (2-2) and (2-3) as a thickener, a base oil consisting of the silicone oil represented by the above-mentioned formula (1), with no other base oil component being added, and magnesium oxide, zinc oxide or molybdenum oxide as a friction modifier.

[0029] In the above embodiment, it is more preferable that the silicone grease composition comprise a mixture of three kinds of diurea compounds represented by the above-mentioned formulas (2-1), (2-2) and (2-3) as the thickener; the base oil consisting of the silicone oil represented by the above-mentioned formula (1) where R1 and R2 are each independently methyl group or phenyl group, with the ratio of methyl group to the whole organic groups being 50 to 100 mol.%, with no other base oil component being added; and magnesium oxide or zinc oxide or molybdenum oxide as the friction modifier, with the amount of the friction modifier being 0.1 to 10 mass% based on the total mass of the composition. [0030] In the above embodiment, it is most preferable that the silicone grease composition comprise a mixture of three kinds of diurea compounds represented by the above-mentioned formulas (2-1), (2-2) and (2-3) as the thickener; the base oil consisting of the silicone oil represented by the above-mentioned formula (1) where R1 and R2 are each independently methyl group or phenyl group, with the ratio of methyl group to the whole organic groups being 95 mol.%, with no other base oil component being added; and magnesium oxide as the friction modifier, with the amount of the friction modifier being 1.0 to 3.0 mass% based on the total mass of the composition.

**[0031]** The silicone grease composition of the invention can be suitably used for the portions to be lubricated where a high coefficient of friction and excellent wear preventive characteristics are needed, to be more specific, the portions of clutch and torque limiter mechanism. More specifically, the silicone grease composition of the invention is usable for the overrunning clutch of automobile starters, the one way clutch of office equipment, a variety of traction driving mechanisms, and the like. Desirably, the surface of the portions to be lubricated may be a member made of steel.

[Examples]

- 50 <Preparation of silicon grease compositions>
  - Base oil

**[0032]** A silicone oil of formula (1) where R1 and R2 are each independently methyl group or phenyl group, with the ratio of methyl group to the whole organic groups being 95 mol.% was used.

- Thickener

5

10

15

20

30

35

40

45

55

[0033] An alicyclic aliphatic diurea prepared from diphenylmethane-4,4'-diisocyanate, cyclohexylamine and octade-cylamine was used.

- Friction modifiers

**[0034]** Magnesium oxide (Mohs hardness: 5-6; Average particle diameter:  $0.5 \mu m$ ) Zinc oxide (Mohs hardness: 4-5; Average particle diameter:  $0.2 \mu m$ ) Molybdenum oxide (Mohs hardness: 2-3; Average particle diameter:  $2.1 \mu m$ ) Titanium oxide (Mohs hardness: 7-8; Average particle diameter:  $0.15 \mu m$ ) Boron nitride (Mohs hardness: 10; Average particle diameter:  $4 \mu m$ )

**[0035]** Predetermined amounts of amines (i.e., cyclohexylamine and octadecylamine) were reacted with diphenylmethane diisocyanate in the base oil to prepare a base grease. By adding the base oil and other additives to the base grease, the resultant mixture was subjected to a milling treatment so as to have a worked penetration of 300 (JIS K2220), thereby obtaining a grease composition.

<Test methods>

- Low-speed and high-load friction test

[0036] In this test, the lubrication conditions of the clutch in the torque-transmitted state were assumed.

**[0037]** The test was conducted using a Falex test machine as prescribed in ASTM D2670. A test grease was applied to the journal and the blocks. A predetermined load was applied to both blocks against the journal in advance, and the test machine rotated the journal for one second. The generated frictional force was recorded, and then the initial coefficient of friction was determined 0.001 sec after starting. The test pieces and the test conditions are as shown below.

Journal: outer diameter 1/4n, SAE 3135 Steel, Rb 87-91

Blocks: AISI 1137 Steel, Rc 20-24 Contact pressure: 200 kgf/mm<sup>2</sup> Circumferential speed: 0.096 m/sec

- High-speed and Low-load abrasion test

[0038] In this test, the lubrication conditions of the clutch in the torque non-transmitted state were assumed.

**[0039]** The test was conducted using a LFW#1 test machine as prescribed in ASTM D2714. A load was applied to a roller against a ring. The test machine rotated the ring to determine the abrasion occurring on the roller using a micrometer. The test pieces and the test conditions are as shown below.

Ring: outer diameter 35 mm x width 8.7 mm, SAE 4620 Steel, Rc 58-63, RMS 6-12

Roller: Cylindrical roller for roller bearing, diameter 6 mm x 6 mm, SUJ-2

Contact pressure: 10 kgf/mm<sup>2</sup> Circumferential speed: 12.8 m/sec

<Evaluation criteria>

- Low-speed and high-load friction test

[0040] Coefficient of friction:

50 oo:  $0.030 \le \mu$ o:  $0.025 \ \mu < 0.030$  $\Delta$ :  $0.020 \ \mu < 0.025$ x:  $\mu < 0.020$ 

- High-speed and low-load abrasion test

[0041] Depth of abrasion

o: < 6 mm

 $x{:} \geq 6 \ mm$ 

10

15

20

25

30

35

40

45

[0042] The results are shown in Tables 1 and 2. In the Tables, the amounts of the thickeners and the friction modifiers are expressed by mass%, which is based on the total mass of each grease composition. The balance is occupied by base oil.

### [Table 1]

		Example 1		Example 2		Example 3		Example 4	
Thickener Type Ratio		Alicyclic aliphatic diurea 15 mass%		Alicyclic aliphatic diurea 14 mass%		Alicyclic aliphatic diurea 15 mass%		Alicyclic aliphatic diurea 15 mass%	
Type of base oil		Methylphenyl silicone		Methylphenyl silicone		Methylphenyl silicone		Methylphenyl silicone	
Friction modifier	Туре	Magnesium oxide, 1 mass%		Magnesium oxide, 3 mass%		Zinc oxide, 3 mass%		Molybdenum oxide, 3 mass%	
	Mohs hardness	5-6		5-6		4-5		2-3	
Coefficient of friction		0.031	00	0.032	00	0.028	0	0.022	Δ
Depth of abrasion (mm)		4.0	0	4.1	0	5.5	0	3.5	0

[Table 2]

[1.0000 =]							
		Comparative Example 1		Comparative Example 2		Comparative Example 3	
Thickener Type	Alicyclic aliphatic diurea		Alicyclic aliphatic diurea		Alicyclic aliphatic diurea		
Ratio	15 mass%		15 mass%		16 mass%		
Base oil Type	Methylphenyl silicone		Methylphenyl silicone		Methylphenyl silicone		
Friction modifier	Туре	Titanium ox mass%	kide 3	Boron nitride 3 mass%			
	Mohs hardness	7-8				-	
Coefficient of friction		0.020	Δ	0.030	00	0.018	х
Depth of abrasion (mm)		15.0	х	7.2	х	4.8	0

### Claims

- 1. A silicone grease composition comprising a thickener; a base oil comprising a silicone oil in an amount of 50 mass% or more of the total mass of the base oil; and a friction modifier comprising a metallic oxide with a Mohs hardness of 6 or less.
- 50 2. The silicone grease composition of claim 1 or 2, wherein the metallic oxide has a Mohs hardness of 2 to 6.
  - **3.** The silicone grease composition of claim 1 or 2, wherein the metallic oxide is magnesium oxide, zinc oxide or molybdenum oxide.
- 55 **4.** The silicone grease composition of any one of claims 1 to 3, wherein the metallic oxide is contained in an amount of 0.1 to 10 mass% based on the total mass of the composition.

#### INTERNATIONAL SEARCH REPORT International application No. PCT/JP2015/054383 CLASSIFICATION OF SUBJECT MATTER 5 C10M169/04(2006.01)i, C10M107/50(2006.01)n, C10M125/10(2006.01)n, C10N10/04(2006.01)n, C10N10/12(2006.01)n, C10N20/00(2006.01)n, C10N30/06 (2006.01)n, C10N40/04(2006.01)n, C10N50/10(2006.01)n According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED 10 Minimum documentation searched (classification system followed by classification symbols) C10M169/04, C10M107/50, C10M125/10, C10N10/04, C10N10/12, C10N20/00, C10N30/06, C10N40/04, C10N50/10 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 15 1971-2015 Toroku Jitsuyo Shinan Koho 1994-2015 Kokai Jitsuyo Shinan Koho Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) 20 DOCUMENTS CONSIDERED TO BE RELEVANT Category\* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Χ JP 9-59664 A (NSK Ltd.), 1 - 404 March 1997 (04.03.1997), claims; examples 25 (Family: none) JP 2004-176075 A (NSK Ltd.), Χ 1 - 424 June 2004 (24.06.2004), claims; examples (Family: none) 30 Χ JP 7-102274 A (Dow Corning Corp.), 1 - 418 April 1995 (18.04.1995), claims; paragraph [0035]; examples & US 6040277 A & EP 648832 A1 35 × See patent family annex. Further documents are listed in the continuation of Box C. 40 Special categories of cited documents: later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "L" 45 document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is document referring to an oral disclosure, use, exhibition or other means combined with one or more other such documents, such combination being obvious to a person skilled in the art document published prior to the international filing date but later than the document member of the same patent family priority date claimed Date of the actual completion of the international search Date of mailing of the international search report 50 27 March 2015 (27.03.15) 07 April 2015 (07.04.15) Name and mailing address of the ISA/ Authorized officer Japan Patent Office 3-4-3, Kasumigaseki, Chiyoda-ku, Tokyo 100-8915, Japan Telephone No. 55 Form PCT/ISA/210 (second sheet) (July 2009)

## INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2015/054383

5	C (Continuation)	). DOCUMENTS CONSIDERED TO BE RELEVANT	101/0120	)15/054383
	Category*	Citation of document, with indication, where appropriate, of the releva	ant passages	Relevant to claim No.
0	X	JP 8-143883 A (Dow Corning Asia, Ltd.), 04 June 1996 (04.06.1996), comparative examples 2, 8 & US 5908815 A & EP 717099 A1		1-4
5	Х	JP 3-106996 A (Hitachi, Ltd.), 07 May 1991 (07.05.1991), claims; page 3, lower left column, 2nd l from the bottom to page 4, upper right column 11; examples (Family: none)	ine olumn,	1-4
0	Х	JP 2012-224834 A (Toyota Motor Corp. et 15 November 2012 (15.11.2012), claims; paragraph [0031]; examples (Family: none)	al.),	1-4
	А	JP 2003-176489 A (NSK Ltd.), 24 June 2003 (24.06.2003), entire text (Family: none)		1-4
5		(1		
5				
5				
,				
5	DCT/IC + O			

Form PCT/ISA/210 (continuation of second sheet) (July 2009)

### 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 HEI735824 B **[0004]**
- JP HEI5230486 A **[0004]**

- JP HEI6279777 A [0004]
- JP 2003176489 A **[0004]**