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(71) Applicant: **WACKER CHEMIE AG** [DE/DE]; Hanns-Seidel-Platz 4, 81737 München (DE).

(72) Inventors: **PARK, Jihye**; c/o Wacker Chemicals Korea Inc., Pangyo Techno Valley, S-3F, 231 Pangyoyeok-ro, Bundang-gu, Seongnam-si, Gyeonggi-do 13494 (KR). **JOO, YoungHyuk**; c/o Wacker Chemicals Korea Inc., Pangyo Techno Valley, S-3F, 231 Pangyoyeok-ro, Bundang-gu, Seongnam-si, Gyeonggi-do 13494 (KR). **KUHN, Arvid**; Am Emetsberger Hof 35, 84489 Burghausen (DE). **YANG, HyunKwan**; c/o Wacker Chemicals Korea Inc., Pangyo Techno Valley, S-3F, 231 Pangyoyeok-ro, Bundang-gu, Seongnam-si, Gyeonggi-do 13494 (KR). **YU, HongJeong**; c/o Wacker Chemicals Korea Inc., Pangyo Techno Valley, S-3F, 231 Pangyoyeok-ro, Bundang-gu, Seongnam-si, Gyeonggi-do 13494 (KR).

(74) Agent: **MIESKES, Klaus** et al.; Wacker Chemie AG, Hanns-Seidel-Platz 4, 81737 München (DE).

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(54) Title: THERMOSETTING SILICONE RESIN COMPOSITION FOR REFLECTOR OF LED, REFLECTOR OF LED AND SEMICONDUCTOR DEVICE USING THE SAME

(57) Abstract: Disclosed is a thermosetting silicone resin composition for a reflector of a Light Emitting Diode (LED), a reflector of a LED and a semiconductor device using the same.

THERMOSETTING SILICONE RESIN COMPOSITION FOR REFLECTOR OF LED, REFLECTOR OF LED AND SEMICONDUCTOR DEVICE USING THE SAME

Technical Field

The present invention relates to a thermosetting resin silicone composition which has high light reflectance, high heat resistance, and low yellowing properties and is used for a light-emitting diode (LED) reflector, and also relates to an LED reflector and a semiconductor device which use the thermosetting silicone composition.

Background Art

In recent years, optical semiconductor devices including optical semiconductor elements such as light-emitting diodes (LEDs) have been used in various applications due to their advantages such as high energy efficiency and long lifespan characteristics, and the demand therefor has increased.

Such optical semiconductor devices are generally supplied in a form in which an optical semiconductor element is mounted on a substrate (optical semiconductor element-mounting substrate) and the optical semiconductor element is sealed with a transparent resin which, if necessary, contains a phosphor. In order to increase the efficiency of light extraction from the optical semiconductor element, a reflection member (reflector) for reflecting light is provided on the substrate.

The reflector is generally manufactured by adding a white pigment to a thermosetting resin such as epoxy resin, followed by transfer molding. This reflector has a problem in that it is yellowed due to increased heat generation resulting from the high brightness of the optical semiconductor device and due to deterioration of the

element material, and thus the light reflectance thereof is significantly reduced.

Therefore, it is an object of the present invention to provide a thermosetting resin composition for an LED reflector, which not only exhibits high light reflectance, but also continuously exhibits heat resistance, low yellowing, and excellent mechanical properties during the driving of an optical semiconductor element and has excellent process suitability applicable to various processes, and to also provide an LED reflector and a semiconductor device which use the thermosetting resin composition.

Disclosure of Invention

The thermosetting silicone resin composition for a reflector of a LED according to the present invention comprises:

(A) a branched organopolysiloxane having at least one silicon-bonded alkenyl group and at least one silicon-bonded aryl group per molecule, and having siloxane units represented by the general formula: $RSiO_{3/2}$ where R is a substituted or unsubstituted monovalent hydrocarbon group;

(B) a linear organopolysiloxane with both terminal ends of the molecular chain blocked by silicon-bonded hydrogen atoms and having at least one silicon-bonded aryl group per molecule;

(C) a hydrosilylation reaction catalyst;

(D) a low molecular weight siloxane having at least one silicon-bonded alkenyl group per molecule, represented by the average formula

$$(R^5{}_3SiO_{1/2})_f(R^5{}_2SiO_{2/2})_g(R^5SiO_{3/2})_h(SiO_{4/2})_i$$

where each R^5 can be the same or different and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^5 per molecule is an alkenyl group, with the proviso that the ratio between alkenyl groups

and silicon atoms is from 0.3 to 1 (as determined by ^{29}Si NMR spectroscopy),

f, g, h, and i are independently 0 or positive, and

the weight average molecular weight Mw of the siloxane is less than 1,000 g/mol (as measured by SEC, THF as solvent, in a concentration of 5 mg/mL, RI detector against polystyrene as standard); and

(E) a white pigment.

In addition, the reflector of LED according to the present invention comprises the above-described thermosetting silicone resin composition.

Furthermore, the semiconductor device of the present invention comprises a reflector cured by using the above-described thermosetting silicone resin composition.

Effects of Invention

The thermosetting silicone resin composition according to the present invention not only exhibits excellent light reflectance, but also rarely undergoes changes in its weight and hardness at high temperatures, indicating that it has excellent heat resistance and mechanical properties. Furthermore, the composition may have excellent process suitability applicable to various processes as a result of suitably controlling the components and their contents in the composition.

Accordingly, the present invention may provide a LED reflector stable against light emitted from an LED chip and heat generated during operation.

In addition, the present invention may improve the reliability of a semiconductor device including the LED reflector because the LED reflector exhibits high light reflectance and hardness at high temperatures and has low yellowing properties.

Detailed Description of the Invention

Hereinafter, an exemplary embodiment of the present invention will be described in detail. However, the exemplary embodiment of the present invention may be modified in various forms, and the scope of the present invention is not limited to an exemplary embodiment to be described below.

<Thermosetting silicone resin composition for a reflector of an LED>

The present invention provides a thermosetting silicone resin composition for a reflector of a LED comprising:

(A) a branched organopolysiloxane having at least one silicon-bonded alkenyl group and at least one silicon-bonded aryl group per molecule, and having siloxane units represented by the general formula: $RSiO_{3/2}$ where R is a substituted or unsubstituted monovalent hydrocarbon group;

(B) a linear organopolysiloxane with both terminal ends of the molecular chain blocked by silicon-bonded hydrogen atoms and having at least one silicon-bonded aryl group per molecule;

(C) a hydrosilylation reaction catalyst;

(D) a low molecular weight siloxane having at least one silicon-bonded alkenyl group represented by the average formula

$$(R^5{}_3SiO_{1/2})_f(R^5{}_2SiO_{2/2})_g(R^5SiO_{3/2})_h(SiO_{4/2})_i$$

where each R^5 can be the same or different and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^5 per molecule is an alkenyl group, with the proviso that the ratio between alkenyl groups and silicon atoms is from 0.3 to 1,

f, g, h, and i are independently 0 or positive,

the weight average molecular weight Mw of the siloxane is less than 1,000 g/mol; and (E) a white pigment.

Hereinafter, a chemical composition of the thermosetting silicone resin composition according to the present invention will be described in detail. However, the present invention is not limited to the following exemplary embodiments, but is embodied in various different forms.

Component (A)

Component (A), which is the major component of the composition according to the present composition, is used to impart strength to the cured product obtained by curing the composition.

Component (A) represents a branched organopolysiloxane having at least one silicon-bonded alkenyl group and at least one silicon-bonded aryl group per molecule, and having siloxane units represented by the general formula: $RSiO_{3/2}$ where R is a substituted or unsubstituted monovalent hydrocarbon group.

In component (A), examples of the alkenyl groups include vinyl, allyl, methallyl, butenyl, pentenyl, and hexenyl group, preferably vinyl and allyl group, particularly preferred vinyl group.

In component (A), examples of the aryl groups include phenyl, naphthyl, anthryl, phenanthryl, indenyl, benzophenyl, fluorenyl, xanthenyl, anthronyl; aryloxyaryl groups such as o- or p-phenoxyphenyl; alkaryl groups such as o-, m-, p-tolyl, xylyl and ethylphenyl; aralkyl groups such as benzyl, α- and β-phenylethyl. Preferably the aryl group is phenyl group.

In addition, examples of silicon-bonded organic groups of component (A) other than the alkenyl and aryl groups include substituted or unsubstituted monovalent

hydrocarbon groups, examples of which include methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, and other alkyl groups; and halogenated alkyl groups such as chloromethyl, 3-chloropropyl, and 3,3,3-trifluoropropyl, with methyl being particularly preferable.

In the siloxane units of component (A) represented by the general formula $RSiO_{3/2}$, R is a substituted or unsubstituted monovalent hydrocarbon group. Substituents of hydrocarbon groups may include the above-mentioned alkyl groups, the above-mentioned alkenyl groups, the above-mentioned aryl groups, the above-mentioned aralkyl groups, and the above-mentioned halogenated alkyl groups, particularly preferably the above-mentioned alkyl groups and the above-mentioned aryl groups.

As component (A), an organopolysiloxane represented by the average unit formula:

$(R^1R^1R^3SiO_{1/2})_a(R^1{}_2SiO_{2/2})_b(R^2SiO_{3/2})_c(SiO_{4/2})_d(XO_{1/2})_e$

is preferable.

In the formula above, each of R^1, R^2, and R^3 can be the same or different and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^1, R^2 or R^3 per molecule is an alkenyl group and at least one of R^1, R^2 or R^3 per molecule is an aryl group.

The monovalent hydrocarbon group may be more specifically exemplified by the above-mentioned alkyl groups, the above-mentioned alkenyl groups, the above-mentioned aryl groups, the above-mentioned aralkyl groups, and the above-mentioned halogenated alkyl groups.

Preferably 0.1 to 40 mol%, more preferably 5 to 25 mol%, of R^1, R^2, and R^3 per molecule is the above-mentioned alkenyl groups. This is due to the fact that when the content of the alkenyl groups is below the lower limit or exceeds the upper limit of the

above-mentioned range, its reactivity with component (A) tends to decrease.

Also, in order to achieve low attenuation due to light refraction, reflection, scattering etc. in the cured product obtained by curing preferably not less than 10 mol% of R^1, R^2, and R^3 should be the above-mentioned aryl groups, And, in particular, in siloxane units represented by the general formula $R^2SiO_{3/2}$, it is even more preferable that not less than 30 mol % of R^2 should be represented by the above-mentioned aryl groups, with R^2 other than the alkenyl and aryl groups being preferably represented by methyl groups.

In addition, in the formula above,

X is a hydrogen atom or alkyl group, a is 0 or a positive number, b is 0 or a positive number, c is a positive number, d is 0 or a positive number, e is 0 or a positive number, b/c is a number between 0 and 10, a/c is a number between 0 and 0.5, d/(a+b+c+d) is a number between 0 and 0.3, and e/(a+b+c+d) is a number between 0 and 0.4.

As component (A), an organopolysiloxane with the average unit formula:

$$(R^1R^1R^3SiO_{1/2})_a(R^1{}_2SiO_{2/2})_b(R^2SiO_{3/2})_c$$

is particularly preferable.

In the formula above, R^1 is a C_1 to C_{12} alkyl group, R^2 is a C_6 to C_{20} aryl group or C_7 to C_{20} aralkyl group, R^3 is C_2 to C_{12} alkenyl group, a is 0 or a positive number, b is 0 or a positive number, and c is a positive number.

Although there are no limitations concerning the molecular weight of component (A), when converted to standard polystyrene, its weight average molecular weight (Mw) should preferably be in the range of from 500 g/mol to 10,000 g/mol, and, especially preferably, in the range of from 700 g/mol to 7,000 g/mol, more preferably in the range from 1,000 g/mol to 5,000 g/mol, particularly from 1,500 to 3,000 g/mol.

Component (A) is preferably present in an amount that equals or is greater than 70 %, more preferably 70 to 90 % by weight, particularly preferred, 75 to 85 % by weight, based on the sum of the amount of components (A) and (B).

According to an implementation of the present invention, the thermosetting silicone resin composition comprises 60 to 75 parts by weight, more preferably 70 to 75 parts by weight, of Component (A), based on the 100 parts by weight of total organosiloxane which is a sum of component (A), component (B), component (C) and component (D).

Component (B)

Component (B) is the curing agent of the present composition.

Component (B) is a linear organopolysiloxane with both terminal ends of the molecular chain blocked by silicon-bonded hydrogen atoms having at least one silicon-bonded aryl group per molecule. By using a linear organopolysiloxane as the curing agent, instead of a branched organopolysiloxane, a good elongation performance may be obtained.

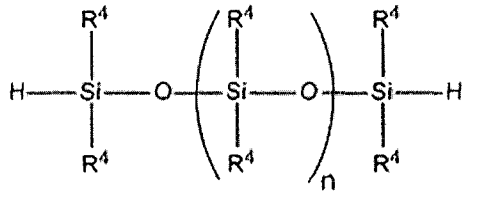
Examples of the aryl groups of component (B) are the same as those described above. Phenyl group is especially preferable.

In addition, examples of silicon-bonded organic groups of component (B) other than the aryl groups include substituted or unsubstituted monovalent hydrocarbon groups with the exception of alkenyl groups, such as the above-described alkyl groups, the above-described aralkyl groups, and the above-described halogenated alkyl groups, with methyl being particularly preferable.

In order to achieve low attenuation due to light refraction, reflection, scattering etc. in the cured product obtained by curing the content of the silicon-bonded aryl groups

among all the silicon-bonded organic groups in component (B) should preferably be not less than 15 mol % and, particularly preferably, not less than 30 mol%. Although there are no limitations concerning the viscosity of component (B) at 25 °C, it is preferably in the range of from 1 to 1,000 mPa·s, and, especially preferably, in the range of from 2 to 500 mPa·s. This is due to the fact that when the viscosity of component (B) is below the lower limit of the above-mentioned range, it may tend to volatilize and the makeup of the resultant composition may be unstable, and, on the other hand, when it exceeds the upper limit of the above-mentioned range, the handling properties of the resultant composition tend to deteriorate.

An organopolysiloxane represented by the general formula:



[Formula 2]

is preferable as component (B).

In the formula above, each R^4 can be the same or different and is independently selected from a hydrogen atom or a substituted or unsubstituted monovalent hydrocarbon group with the exception of alkenyl groups.

Examples of the monovalent hydrocarbon groups of R^4 include the above-mentioned alkyl groups, the above-mentioned aryl groups, and the above-mentioned halogenated alkyl groups.

Here, at least one R^4 per molecule must be one of the above-mentioned aryl groups, preferably, phenyl.

In addition, n in the formula above is an integer of 0 or more, preferably, an integer in the range of from 0 to 20, and, especially preferably, an integer in the range of

from 0 to 10. This is due to the fact that when the value of n exceeds the upper limit of the above-mentioned range, the toughness of the resultant composition, or the adhesive properties of the cured product, tend to deteriorate. It is most preferable that n is 1 to 4, particularly, 1, that is, the component (B) represents trisiloxane.

By having M unit as the repeating unit, instead of Q or T unit, a good elongation performance may be obtained.

Component (B) is preferably present in an amount of 1 to 30 % by weight, more preferably 10 to 30 % by weight, particularly prefered, 15 to 25 % by weight, based on the sum of the amount of components (A) and (B).

It is preferable that the molar ratio of Si-H groups in component (B)/ alkenyl groups, for example, vinyl groups, in component (A) is 1 to 1.2 to reduce the reactive residual silicone hydride.

According to an implementation of the present invention, the thermosetting silicone resin composition comprises 18 to 25 parts by weight, more preferably 20 to 22 parts by weight, of Component (B), based on the 100 parts by weight of total organosiloxane which is a sum of component (A), component (B), component (C) and component (D).

Component (C)

Component (C) is a hydrosilylation reaction catalyst.

The hydrosilylation reaction catalyst of component (C) is used to promote the reaction of the alkenyl groups of component (A) with the silicon-bonded hydrogen atoms of component (B).

Examples of component (C) include platinum catalysts, rhodium catalysts, and palladium catalysts. Platinum catalysts are preferable because of their ability to

significantly stimulate the cure of the present composition. Examples of the platinum catalysts include platinum micropowder, chloroplatinic acid, alcohol solutions of chloroplatinic acid, platinum/alkenylsiloxane complexes, platinum/olefin complexes, and platinum/carbonyl complexes, preferably, platinum/alkenylsiloxane complexes. Examples of the alkenylsiloxanes include 1,3-divinyl-1,1,3,3-tetramethyldisiloxane, 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane, alkenylsiloxanes obtained by substituting groups such as ethyl, phenyl etc. for some of the methyl groups of the above-mentioned alkenylsiloxanes, and alkenylsiloxanes obtained by substituting groups such as allyl, hexenyl, etc. for the vinyl groups of the above-mentioned alkenylsiloxanes. 1,3-divinyl-1,1,3,3-tetramethyldisiloxane is particularly preferable because of the excellent stability of the platinum/alkenylsiloxane complex. Also, due to the improvement in the stability of the complex that their addition may bring, it is desirable to add 1,3-divinyl-1,1,3,3-tetramethyldisiloxane, 1,3-diallyl-1,1,3,3-tetramethyldisiloxane, 1,3-divinyl-1,3-dimethyl-1,3-diphenyldisiloxane, 1,3-divinyl-1,1,3,3-tetraphenyldisiloxane, 1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane and other alkenylsiloxanes and organosiloxane oligomers such as dimethylsiloxane oligomers to the platinum/alkenylsiloxane complex, with alkenylsiloxanes being particularly preferable.

There are no limitations on the content of component (C) as long as the amount promotes curing of the present composition. However, specifically, in the present composition, component (C) is preferably present in an amount resulting in a platinum content of 0.05 to 100 ppm (parts per million) by weight, more preferably 0.1 to 10 ppm by weight, particularly preferred 0.1 to 5 ppm by weight, relative to 100 parts by weight of the total of components (A) and (B). This is due to the fact that when the content of component (C) is below the lower limit of the above-mentioned range, the present

composition tends to fail to completely cure, and, on the other hand, when it exceeds the upper limit of the above-mentioned range, problems may arise in terms imparting various colors to the resultant cured product.

Component (D)

Component (D) is an additive, which is a low molecular weight siloxane having at least one silicon-bonded alkenyl group per molecule, represented by the average formula

$$(R^5{}_3SiO_{1/2})_f(R^5{}_2SiO_{2/2})_g(R^5SiO_{3/2})_h(SiO_{4/2})_i$$

where each R^5 can be the same or different and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^5 per molecule is an alkenyl group, with the proviso that the ratio between alkenyl groups and silicon atoms is from 0.3 to 1,

f, g, h, and i are independently 0 or positive, and

the weight average molecular weight Mw of the siloxane is less than 1,000 g/mol, preferably less than 800 g/mol, more preferably less than 500 g/mol.

Preferably component (D) is selected from the group consisting of $D^{alkenyl}{}_4$, $M^{alkenyl}{}_4Q$, $M^{alkenyl}{}_6Q_2$, $M^{alkenyl}{}_3T$. Preferably the alkenyl group is vinyl. Particularly preferred as component (D) are cyclotetrasiloxane $D^{Vi}{}_4$, and $M^{Vi}{}_4Q$, especially $D^{Vi}{}_4$.

It is assumed that component (D) serves to react with an unreacted site (Si-H), thereby protecting additional reaction of the unreacted site due to heat or light. As a result, in the composition comprising component (D), color change and mechanical strength change to high temperature may be remarkably reduced.

In the composition according to prior art, which has no component (D), materials often show an increase in hardness after curing during storage at high temperature,

which can be attributed to a post-curing reaction of remaining reactive sites and/or show a weight loss by evaporation of low molecular weight species that did not crosslink into the polymer network. Due to the change in hardness and weight loss, the materials were not stable under operating conditions. In the present invention, a reaction of component (D) with unreacted site (Si-H) leads to better stability such as less change in hardness, low weight loss, and discoloration, at high temperature.

Component (D) also gives a filling effect, which makes it possible to lower gas and vapor transmission. The encapsulant further comprising component (D) provides stablity to sulfur and such effect is expected to be obtained from the filing effect.

As a result, an LED package with excellent reliability may be obtained.

Component (D) is both reactive and small enough to react with residual silicone hydride. At the same time, component (D) crosslinks into the siloxane network and thus does not increase the proportion of volatile components in the cured material. Component (D) is preferably used in an amount of at most 10 parts by weight, more preferably at most 7 parts by weight, and even more preferably at most 5 parts by weight relative to 100 parts by weight of the total of components (A) and (B). If component (D) is included in higher amounts, compatibility with the other components would be lower and thus, transmittance would be lower. Preferably, component (D) is used in an amount of at least 1 parts by weight relative to 100 parts by weight of the total of components (A) and (B). The amount of the component (D) may be adequately selected in consideration of the specific formulation and the feature.

According to an implementation of the present invention, the thermosetting silicone resin composition comprises 1 to 5 parts by weight, more preferably 2 to 4 parts by weight, of Component (D), based on the 100 parts by weight of total organosiloxane which is a sum of component (A), component (B), component (C) and component (D).

Component (E)

Component (E) is a white pigment.

The white pigment (E) functions to impart high light reflectance to a reflector, which is the cured product of the composition, and also to reduce the linear expansion of the reflector.

The white pigment (E) may be any white pigment known in the art, for example, an inorganic white pigment, an organic white pigment, a hollow particle pigment, or a mixture of two or more thereof.

Non-limiting examples of the inorganic white pigment include glass, clay, mica, talc, kaolinite (kaolin), halloysite, zeolite, acid clay, activated clay, boehmite, pseudo-boehmite, inorganic oxides; metal salts (e.g., alkaline earth metal salts, etc.), and the like. Furthermore, non-limiting examples of the organic white pigment include resin pigments, such as styrene-based resins, benzoguanamine-based resins, urea-formalin resins, melamine-formalin resins, amide-based resins, and the like.

Preferably, the white pigment (E) in the present invention, may be at least one selected from the group consisting of titanium oxide, zinc oxide, silicon oxide, aluminum oxide, barium oxide, magnesium oxide, zirconium oxide, aluminum silicate, boron nitride, calcium carbonate, tantalum pentoxide, barium sulfate, magnesium carbonate, barium carbonate, and strontium titanate.

The white pigment (E) preferably has high refractive index in order to increase the reflectance of the reflector. In one example, a white pigment having a refractive index of 1.5 or higher is preferable. In this case, a white pigment having a hollow particle structure has a very high surface reflectance due to low refractive index gas contained in the core, ands for this reason, the shell portion may also be composed of a

material having a refractive index lower than 1.5. In addition, the pigment white (E) may be a pigment subjected to surface treatment known in the art. In one example, the surface treatment comprises surface treatment with a surface treatment agent, such as a metal oxide, a silane coupling agent, a titanium coupling agent, an organic acid, a polyol, silicone or the like. When this surface treatment is performed, it may improve either the compatibility of the white pigment with other components, included in the silicone resin composition according to the present invention, or the dispersibility of the white pigment.

In addition, the shape of the white pigment (E) is not particularly limited, and may be, for example, a spherical shape, a crushed shape, a fibrous shape, a needle shape, a scaly shape, an amorphous shape, or the like. From a viewpoint of dispersibility, a spherical shape is preferably, and in particular, a spherical shape having an aspect ratio of 1.2 or less is more preferable.

In addition, the mean particle diameter of the white pigment is not particularly limited, but may be 0.01 to 50 μm, preferably 0.1 to 50 μm, in view of increasing the light reflectance of the curing product (white reflector). As used herein, the term "average diameter" means the diameter at 50% (median diameter, D_{50}) of the cumulative particle size distribution measured by a laser diffraction/scattering method.

The content of the white pigment (E) in the thermosetting silicone resin composition according to the present invention is not particularly limited and may be suitably adjusted within the range known in the art. In one example, the content of the white pigment (E) may be 1 to 300 parts by weight, preferably 20 to 250 parts by weight, more preferably 50 to 200 parts by weight, based on 100 parts by weight of the sum of component (A) to component (D). When the content of the white pigment is within the above-described range, the light reflectively of a reflector, which is the

curing product of the composition, can further be increased, and the reduction in flowability of the thermosetting silicone resin composition by addition of the white pigment (E) can also be inhibited, resulting in improvement in workability and moldability.

In one preferable example of the present invention, the white pigment (E) comprises a titanium oxide-based first white pigment (E-1) having a mean particle diameter (D_{50}) of 0.1 to 2 μm. This titanium oxide is preferable in view of increasing the reflectance of the curing product (white reflector) and increasing light reflectance per amount of white pigment added. The titanium oxide may be any titanium oxide known in the art, and may be, for example, rutile-type titanium oxide, anatase-type titanium oxide, brookite-type titanium oxide, or a mixture of two or more thereof.

The content of the first white pigment (E-1) is not particularly limited, but may be at least 40 parts by weight, preferably 45 to 80 parts by weight, based on 100 parts by weight of the sum of component (A) to component (D). When the composition comprises the titanium oxide-based first white pigment having the above-specified particle diameter in an amount of at least 40 parts by weight based on the total weight of component (A) to component (D), it may exhibit a light reflectance of 97% or higher, thereby maximizing the optical efficiency of the LED reflector.

Meanwhile, in the present invention, the viscosity, thixotropy, hardness after heat curing, reflectance, whiteness and the like of the composition, which are required in various manufacturing processes, may be suitably adjusted by using the above-described titanium oxide-based first white pigment (E-1) in combination with pigments having various sizes and components. Accordingly, the composition of the present invention may further comprise a second white pigment (E-2), a third white pigment (E-

3), and a mixture thereof, from which at least one of the mean particle diameter and component of the first white pigment (E-1) differs.

In one preferable example of the present invention, the ratio of the content of the first white pigment (E-1) to the content of the other pigment (e.g., E-2, E-3, E-2+E-3, etc.) except the first white pigment may be 80-99.9: 0.1-20 by weight, preferably 80-99.5: 0.5-20 by weight, based on 100 parts by weight of the total weight of the white pigment (E).

The heat curable silicone composition of the present invention may not comprise as flexibilizing units, a linear organopolysiloxane having at least two silicon-bonded alkenyl groups and at least one silicon-bonded aryl group per molecule. It is assumed that since bulky branched organopolysiloxane such as component (A) is used as resin instead of a linear organopolysiloxane, the present curable silicone composition provides excellent hardness and toughness. That is, bulky branched organopolysiloxane is directly connected to unreacted Si-H site.

The thermosetting silicone resin composition of the present invention may further comprise a crosslinking agent, which is generally used in this field.

The thermosetting silicone resin composition of the present invention may further comprise a curing inhibitor; silica, glass, quartz, cristobalit, alumina, zinc oxide and other inorganic fillers; a catalyst; and a phosphor, which are generally used in this field.

The present composition may also contain; micropowders of organic resins such as polymethacrylate resin; heat-stabilizers, dyes, pigments, flame retardants, solvents, etc. as optional components, so long as this does not impair the purpose of this invention.

The compositions described above may be prepared by mixing the components generally used in this art, for example, by mixing all the components at ambient

temperature. Specifically, the composition may be prepared by mixing the above-described components (A, B, C, D and E) and other additives at a predetermined ratio, uniformly mixing the mixture by a Henschel mixer, a V blender, a ribbon blender, a tumbler blender or the like, followed by melt-kneading using a roll mill, a kneader, an extruder, a Banbury mixer or the like, cooling and solidifying the melt-kneaded material, and then grinding the solid material to a suitable size.

In general, in a process of manufacturing an LED reflector, the viscosity of a composition that may be applied to a dispensing process is 500 to 10,000 mPa·s, and the viscosity of a composition that may be applied to transfer molding and compression molding is 3,000 to 100,000 mPa·s. Since the thermosetting silicone resin composition according to the present invention has a viscosity of 100 to 2,000,000 mPa·s (at 25°C) before curing, it may be applied to all the above-described dispensing process and transfer molding and compression molding processes. In addition, when the thermosetting silicone resin composition is molded by any one of transfer molding and compression molding, it may be cured rapidly at a temperature of 100 to 170°C within 30 to 600 seconds.

As described above, the thermosetting silicone resin composition of the present invention may be applied to various processes, and may be cured rapidly due to high curing rate, indicating that it has excellent workability and moldability. In addition, it has high mechanical strength, heat resistance and light reflectance, and thus may preferably be used for an LED reflector. In addition to its use for the LED reflector, the thermosetting silicone resin composition of the present invention may be used without limitation in various light-emitting applications which require high light reflectance and excellent heat resistance. In one example, it may be used as a reflection member for various light-emitting devices, including electrical/electronic parts, indoor lights, ceiling

lights, outdoor lights, vehicle lights, display devices, headlights, and the like.

<LED reflector>

The LED reflector according to the present invention includes the curing product of the above-described thermosetting silicone resin composition.

The shape or size of the reflector is not particularly limited, and the reflector may include a conventional configuration known in the art. In one example, the reflector may have either a shape opened in at least side in a light emitting direction, or a non-opened planar shape.

The reflector according to the present invention may be manufactured by a conventional molding method. In one example, it is obtained by shaping the above-described thermosetting silicone resin composition into a desired shape by various molding methods, such as transfer molding, compression molding, injection molding, LIM molding (injection molding), blow molding, insert molding, dam molding with a dispenser, etc. In this case, molding and curing of the composition may occur at the same time. Alternatively, a post-curing process may also be separately performed after the molding process. Among these molding methods, the transfer molding or compression molding method is preferably used.

In one specific example, the transfer molding process is preferably performed using a transfer molding machine at a molding pressure of 5 to 20 N/mm^2 at a molding temperature of 120 to 190°C, particularly 120 to 180°C, for 30 to 500 seconds, particularly 30 to 300 seconds.

In one specific example, the compressing molding process is preferably performed using a compression molding machine at a molding temperature of 120 to 190°C, particularly 120 to 180°C, for 30 to 600 seconds, particularly 120 to 420 seconds.

After the above-described molding process, a (post) curing process may, if necessary, be performed. Curing conditions for this process are not particularly limited, and may be suitably adjusted within the range known in the art. In one example, the curing process may be performed at a temperature of 100 to 170°C for 30 seconds to 30 minutes.

The reflector of the present invention, manufactured as described above, may exhibit high reflectance, excellent mechanical properties and low yellowing properties even at high temperature. In one preferable example, the LED reflector according to the present invention may have: (i) a Shore D hardness of at least 10, preferably at least 30, more preferably at least 50; (ii) a light reflectance at 450 nm wavelength of at least 90%, preferably at least 95%, more preferably at least 97%; and (iii) a yellow index of 5 or less, preferably 3 or less.

In the present invention, an optical semiconductor element (e.g., a light-emitting diode element) having the reflector may be fabricated by mounting a conventional LED element and other components, known in the art, on the above-described reflector, and then performing sealing, joining, bonding or the like using an encapsulating resin. This optical semiconductor element (LED assembly) may be used in various applications. In one example, it may be used in applications, including semiconductor devices, LCD displays, backlight units (BLU), mobile phones, vehicle display lights, vehicle head lamps, flashlights, indoor lights, traffic lights, streetlights and outdoor lights.

<Semiconductor device>

The present invention provides a semiconductor device including a reflector obtained by molding the above-described thermosetting silicone resin composition.

As used herein, the term "semiconductor device" includes all semiconductor

devices known in the art. Specifically, the semiconductor device may comprise: the above-described reflector (or a reflector substrate); an optical semiconductor element mounted on the reflector; and a phosphor-containing transparent encapsulating resin layer sealing the optical semiconductor element. The optical semiconductor element may be an LED.

Such semiconductor elements are exemplified by semiconductor elements used in diodes, transistors, thyristors, solid-state image pickup elements, monolithic ICs and in hydride ICs. In particular, it is preferable that semiconductor elements are light-emitting elements.

Examples of such semiconductor devices included diodes, light-emitting diodes, transistors, thyristors, photocouplers, CCDs, monolithic IICs, hybrid ICs, LSIs, and VLSIs.

In the present invention, the LED reflector obtained by molding the above-described thermosetting silicone resin composition exhibits high light reflectance and mechanical properties, and also exhibits excellent heat resistance and low yellowing properties during driving, and thus light-induced deterioration of the reflector surface is minimized. Accordingly, the reliability of an optical semiconductor device (LED device) comprising the LED reflector may be improved.

The present invention will be described in further detail below with reference to examples. However, these examples are intended to illustrate the present invention, but the scope of the present invention is not limited to these examples.

Examples

The invention will now be illustrated by examples, which are not to be construed as limiting the invention in any way.

Analytical Methods

Description of ^{29}Si-NMR measurement:

Solvent: C_6D_6 99,8%d/CCl_4 1:1 v/v with 1 % w/w $Cr(acac)_3$ as reagent for relaxation

Sample concentration: ca. 2 g / 1.5 mL solvent in 10 mm NMR tube

Spectrometer: Bruker Avance 300

Sample head: 10 mm $^{1}H/^{13}C/^{15}N/^{29}Si$ glassfree QNP-Head (Bruker)

Measurement parameter: Pulprog = zgig60, TD = 64k, NS = 1024, SW = 200 ppm, AQ = 2.75 s, D1 = 4 s, SFO1 = 300.13 MHz, O1 = -50 ppm

Processing-Parameter: SI = 64k, WDW = EM, LB = 0.3 Hz

Viscosity:

Viscosity data is measured with a rheometer model MCR302 manufactured by the company Anton Paar, D-Ostfildern, according to DIN EN ISO 3219 in rotation with a cone-plate measurement system. Measurements were performed in a range where the samples behavior is newtonian. Viscosity data are given for a temperature of 25 °C and an ambient pressure of 1013 mbar.

Molecular weight:

Molecular weight is determined as weight average molecular weight Mw and number average molecular Mn by Size Exclusion Chromatography SEC. Polystyrene is used as standard. The detector is a RI detector. THF is used as solvent. Sample concentration is 5 mg / mL.

Synthesis example 1

700 g (2.91 mole) phenyltriethoxysilane, 61.6 g (0.415 mole) diethoxydimethylsilane and 77.6 g (0.416 mole) 1,3-divinyl-1,1,3,3-tetramethyldisiloxane were mixed in a 2 L

round-bottomed flask. 600 g distilled water and 3.00 g 20 % hydrochloric acid were added to the solution. The reaction mixture was refluxed for 2 hours. After cooling, 4.50 g 25 % sodium hydroxide solution were added and the reaction mixture was refluxed for 1 hour. The homogeneous mixture was neutralized with 2.00 g 20 % hydrochloric acid. Ethanol was distilled at 40 °C under vacuum and 1 L ethylacetate and 50 g sodium chloride were added. The aqueous phase was removed and the organic phase was washed three times with saturated aqueous sodium chloride solution. The organic phase was dried with magnesium sulfate and was filtrated with press filter equipment. After removal of the solvent in vacuum, 470 g of a colorless, highly viscous product were obtained. Weight average molecular weight Mw is 2,546 g/mol. The results of ^{29}Si NMR are $ViMe_2SiO_{1/2}$: 16.6 %, $Me_2SiO_{2/2}$: 9.7 %, $Ph(OR)SiO_{2/2}$: 12. 8 % and $PhSiO_{3/2}$: 60.9 %.

Example 1

75 % by weight of the compound from synthesis example 1as component A, 25 % by weight of 1,1,5,5-tetramethyl-3,3-diphenyltrisiloxane as component B were prepared. In addition, relative to 100 parts by weight of the total of components A and B, 0.0002 parts (with respect to Platinum) by weight of Platinum(0)-1,3-divinyl-1,1,3,3,-tetramethyl-disiloxane complex as component C, and 4.99 parts by weight of D^{Vi}_4 as component D were prepared.

Based on 100 parts by weight of the sum of the components (A, B, C, and D), 50 parts by weight of titanium oxide (first white pigment), having a mean particle diameter of 0.3 μm, and 0.5 parts by weight of silicon oxide (second white pigment) having a mean particle diameter of 0.02 μm were prepared as a white pigment (E) and mixed

with the above components, thereby preparing a thermosetting silicone resin composition for an LED reflector according to Example 1.

Examples 2 to 11

The thermosetting silicone resin compositions for reflector of LED according to examples 2 to 11 were prepared in the same manner in example 1 except that component E was used in an amount as shown in following table 1.

[Table 1]

		Examples										
		1	2	3	4	5	6	7	8	9	10	11
Thermosetting silicone composition (A)+(B)+(C)+(D)		100	100	100	100	100	100	100	100	100	100	100
Component (E)	Titanium dioxide (0.3 μm)	50	60	70	50	50	50	50	50	50	50	50
	Silicon oxide (0.02 μm)	0.5	0.5	0.5	-	-	-	-	-	0.5	0.5	0.5
	Boron nitride (3 μm)	-	-	-	10	-	-	-	-	-	-	-
	Boron nitride (5 μm)	-	-	-	-	10	-	-	-	-	-	-
	Boron nitride (30 μm)	-	-	-	-	-	10	-	-	-	-	-
	Aluminum silicate (1 μm)	-	-	-	-	-	-	10	-	-	-	-
	Aluminum silicate (2 μm)	-	-	-	-	-	-	-	10	-	-	-
	Tantalum pentoxide (0.6 μm)	-	-	-	-	-	-	-	-	10	-	-
	Silicon oxide (0.5 μm)	-	-	-	-	-	-	-	-	-	10	-
	Zirconium Oxide (1 μm)	-	-	-	-	-	-	-	-	-	-	10

*unit: parts by weight relative to the 100 parts by weight of a sum of components (A), (B), (C) and (D)

[Evaluation Example]

The physical properties of the thermosetting silicone resin compositions for LED

reflectors, prepared in Examples 1 to 11, were measured as described below, and the results of the measurement are shown in Table 2 below. Molding of the curing products obtained by curing the compositions was performed using a transfer molding machine under the conditions of molding temperature of 165°C, molding pressure of 7 MPa and molding time of 300 seconds. The cured samples prepared by the transfer molding were post-cured at 150°C for about 4 hours.

<Results of evaluation of physical properties>

(1) Measurement of viscosity

For the thermosetting silicone resin composition prepared in each of the Examples, the dynamic viscosities at shear rates of 1/sec and 10/sec were measured using a 25-mm-diameter flat plate in a rheometer model MCR302 (Anton Paar) system.

(2) Thixotropic index

Thixotropic index was calculated as the ratio of the dynamic viscosity at a shear rate of 10/sec to the dynamic viscosity at a shear rate of 1/sec.

(3) Shore hardness (D)

The hardness of a molded sample having a thickness of 6 mm was measured on a Shore Durometer Hardness (D) scale.

(4) Yellow index (b*)

The yellow index of a molded sample having a thickness of 1 mm was measured by CIE Lab colorimetry using Minolta CM5.

(5) Light reflectance

The light reflectance at 450 nm of a transfer-molded sample having a thickness of 1 mm was measured using a Minolta CM 5 spectrophotometer in specular component-included mode.

As shown in Table 2, it could be seen that the compositions of Examples 1 to 11 according to the present invention had high light reflectance, excellent mechanical properties, and low yellowing properties, which are required for a reflector.

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[Table 2]

		Examples										
		1	2	3	4	5	6	7	8	9	10	11
Viscosity [mPa·s] 10/sec, at 25°C	1/sec	15800	23600	32700	43800	25200	22600	15800	18600	20100	19600	19000
	10/sec	4350	5790	7370	7700	6040	6250	5130	5130	5290	5140	4930
Thixotropic Index	ηl1/sec/ηl10/sec	3.63	4.08	4.44	5.69	4.17	3.62	3.08	3.63	3.80	3.81	3.85
Hardness Shore D	6T	76	78	76	76	77	76	80	81	77	78	78
Yellow Index [b*]	1T	2.55	2.59	2.47	2.61	2.39	2.55	3.08	2.44	2.51	2.50	2.89
Reflectance [450nm]	1T	97.14	97.08	97.39	96.31	95.27	96.21	95.54	95.05	97.32	97.22	94.64

WHAT IS CLAIMED IS:

1. A thermosetting silicone resin composition for reflector of LED comprising:

(A) a branched organopolysiloxane having at least one silicon-bonded alkenyl group and at least one silicon-bonded aryl group per molecule, and having siloxane units represented by the general formula: $RSiO_{3/2}$ where R is a substituted or unsubstituted monovalent hydrocarbon group;

(B) a linear organopolysiloxane with both terminal ends of the molecular chain blocked by silicon-bonded hydrogen atoms and having at least one silicon-bonded aryl group per molecule;

(C) a hydrosilylation reaction catalyst;

(D) a low molecular weight siloxane having at least one silicon-bonded alkenyl group represented by the average formula

$$(R^5{}_3SiO_{1/2})_f(R^5{}_2SiO_{2/2})_g(R^5SiO_{3/2})_h(SiO_{4/2})_i$$

where each R^5 can be the same or different and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^5 per molecule is an alkenyl group, with the proviso that the ratio between alkenyl groups and silicon atoms is from 0.3 to 1,

f, g, h, and i are independently 0 or positive,

the weight average molecular weight Mw of the siloxane is less than 1,000 g/mol; and

(E) a white pigment,

wherein the white pigment (E) is contained in an amount of 1 to 300 parts by weight based on 100 parts by weight of a sum of component (A) to component (D).

2. The thermosetting silicone resin composition of claim 1, wherein the composition has a viscosity of 100 to 2,000,000 mPa·s (at 25°C) before curing, and is cured rapidly at a temperature of 100 to 170°C within 30 to 600 seconds when being molded by any one method selected from among transfer molding and compression molding.

3. The thermosetting silicone resin composition of claim 1, wherein the white pigment (E) is at least one selected from the group consisting of titanium oxide, zinc oxide, silicon oxide, aluminum oxide, barium oxide, magnesium oxide, zirconium oxide, aluminum silicate, boron nitride, calcium carbonate, tantalum pentoxide, barium sulfate, magnesium carbonate, barium carbonate, and strontium titanate.

4. The thermosetting silicone resin composition of claim 1, wherein the white pigment (E) has a mean particle diameter (D_{50}) of 0.01 to 50 μm.

5. The thermosetting silicone resin composition of claim 1, wherein the white pigment (E) comprises a titanium oxide-based first white pigment (E-1) having a mean particle diameter of 0.1 to 2 μm.

6. The thermosetting silicone resin composition of claim 5, wherein the first white pigment (E-1) is contained in an amount of at least 40 parts by weight based on 100 parts by weight of a sum of component (A) to component (D).

7. The thermosetting silicone resin composition of claim 5, wherein the white pigment further comprises a second white pigment (E-2), a third white pigment (E-3), or

a mixture thereof, in which at least one of the mean particle diameter and component of the first white pigment (E-1) differs.

8. The thermosetting silicone resin composition of claim 7, wherein a ratio of a content of the first white pigment (E-1) to a content of the remaining pigment exclusive of the first white pigment is 80-99.9: 0.1-20 by weight, based on 100 parts by weight of a total weight of the white pigment (E).

9. The thermosetting silicone resin composition for reflector of LED according to claim 1, in which component (A) is an organopolysiloxane represented by the average unit formula:

$$(R^1R^1R^3SiO_{1/2})_a(R^1{}_2SiO_{2/2})_b(R^2SiO_{3/2})_c(SiO_{4/2})_d(XO_{1/2})_e$$

where each of R^1, R^2, and R^3 can be the same or different, and is independently selected from a substituted or unsubstituted monovalent hydrocarbon group, wherein at least one of R^1, R^2, or R^3 per molecule is an alkenyl group and at least one of R^1, R^2, or R^3 per molecule is an aryl group,

X is a hydrogen atom or alkyl group,

a is 0 or a positive number,

b is 0 or a positive number,

c is a positive number,

d is 0 or a positive number,

e is 0 or a positive number,

b/c is a number between 0 and 10,

a/c is a number between 0 and 0.5,

d/(a+b+c+d) is a number between 0 and 0.3, and

e/(a+b+c+d) is a number between 0 and 0.4.

10. The thermosetting silicone resin composition for reflector of LED according to claim 1, in which component (A) is an organopolysiloxane with the average unit formula:

$$(R^1R^1R^3SiO_{1/2})_a(R^1{}_2SiO_{2/2})_b(R^2SiO_{3/2})_c$$

where R^1 is a C_1 to C_{12} alkyl group,

R^2 is a C_6 to C_{20} aryl group or C_7 to C_{20} aralkyl group,

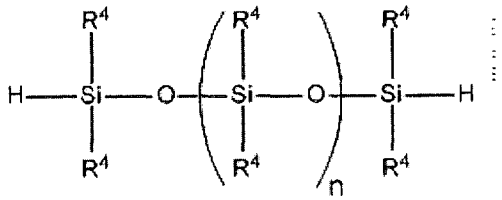
R^3 is C_2 to C_{12} alkenyl group,

a is 0 or a positive number,

b is 0 or a positive number, and

c is a positive number.

11. The thermosetting silicone resin composition for reflector of LED according to claim 1, in which component (B) is an organopolysiloxane represented by the general formula:



[Formula 2]

where each R4 can be the same or different and is independently selected from a hydrogen atom or a substituted or unsubstituted monovalent hydrocarbon group with the exception of alkenyl groups, wherein at least one R4 per molecule is an aryl group, and n is an integer of 0 or more.

12. The thermosetting silicone resin composition for reflector of LED according to claim 1, wherein a weight ratio of component (A) to component (B) is 70:30 to 90:10.

13. The thermosetting silicone resin composition for reflector of LED according to claim 12, wherein a weight ratio of component (A) to component (B) is 75:25 to 85:15.

14. The thermosetting silicone resin composition for reflector of LED according to claim 1, in which component (D) is selected from the group consisting of D^{Vi}_4, M^{Vi}_4Q, $M^{Vi}_6Q_2$, and M^{Vi}_3T.

15. The thermosetting silicone resin composition for reflector of LED according to claim 14, in which component (D) comprises D^{Vi}_4.

16. The thermosetting silicone resin composition for reflector of LED according to claim 1, wherein component (D) is comprised in an amount of at most 10 parts by weight, relative to 100 parts by weight of the total of components (A) and (B).

17. The thermosetting silicone resin composition for reflector of LED according to claim 1, wherein the molar ratio of the hydrosilyl group in component (B) with respect to the alkenyl group in component (A) is 1 to 1. 2.

18. The thermosetting silicone resin composition for reflector of LED according to claim 1, further comprising at least one selected from the group consisting of crosslinking agent, inorganic filler, a curing inhibitor, a catalyst, and a phosphor.

19. An LED reflector comprising a cured product formed by curing the thermosetting silicone resin composition claimed in any one of claims 1 to 18.

20. The LED reflector of claim 17, which has:

a Shore D hardness of 10 or more;

a light reflectance of 90% or higher at a wavelength of 450 nm; and

a yellow index of 5 or less.

21. A semiconductor device comprising the reflector of claim 18.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2018/073484

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09D183/04 C08G77/12 C08G77/20 C08L83/04 H01L23/31
H01L33/56 C08G77/00
ADD.
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08K C08G H01L C08L C09D

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

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2013/115415 A1 (DOW CORNING TORAY CO LTD [JP]) 8 August 2013 (2013-08-08) page 2, paragraph 6 - page 3, paragraph 8 page 5, paragraph 15 example 3 -----	1-21
Y	WO 2018/028792 A1 (WACKER CHEMIE AG [DE]) 15 February 2018 (2018-02-15) page 4, lines 7-19 page 5, lines 7-25 page 15, lines 3-9 claims 1-15 -----	1-21
Y	EP 2 075 280 A1 (SHINETSU CHEMICAL CO [JP]) 1 July 2009 (2009-07-01) page 2, paragraph 9-10 page 5, paragraph 33-37 example 2; table 1 -----	1-21

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
15 March 2019	25/03/2019

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Denis, Cécile

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
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