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(54) **Title**: ROOM-TEMPERATURE-CURABLE ORGANOPOLYSILOXANE COMPOSITION

(57) **Abstract**: A room-temperature-curable organopolysiloxane composition comprising: (A) a diorganopolysiloxane that has a viscosity of 100 to 1,000,000 mPa•s at 25 °C and molecular terminal groups selected from the group consisting of alkoxysilyl groups, hydrogen atoms, hydroxyl groups, and substituted or unsubstituted univalent hydrocarbon groups but where at least 50 mole % of the terminal groups are said alkoxysilyl groups, which are represented by the following formula: $-X-SiR^1_a(OR^2)_{(3-a)}$ (wherein R^1 represents substituted or unsubstituted univalent hydrocarbon groups, R^2 represents alkyl groups, X represents oxygen atoms or alkylene groups, and "a" is 0 or 1); (B) an organotrimethoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrimethoxysilane: $R^3Si(OCH_3)_3$ (wherein R^3 represents substituted or unsubstituted univalent hydrocarbon groups); (C) an organotrialkoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrialkoxysilane: $R^4Si(OR^5)_3$(wherein R^4 represents substituted or unsubstituted univalent hydrocarbon groups, and R^5 represents alkyl groups with two or more carbon atoms); and (D) a titanium chelate catalyst. The composition possesses excellent storage stability in a moisture-free environment and provides easy control of a curing speed.

DESCRIPTION

ROOM-TEMPERATURE-CURABLE ORGANOPOLYSILOXANE COMPOSITION

Technical Field

[0001] The present invention relates to a room-temperature-curable organopolysiloxane composition and, more specifically, to a room-temperature-curable organopolysiloxane composition that shows excellent stability under moisture-free conditions and allows easy control of a curing speed.

Background Art

[0002] It is known that some specific organopolysiloxane compositions, which are curable at room temperature under the effect of moisture of air (see Japanese Unexamined Patent Application Publication S62-252456), show excellent storage stability under moisture-free conditions and are suitable for forming cured products that demonstrate excellent adhesiveness in humid air, provide easy evaporation of methanol, ethanol, or similar alcohols when cured by a de-alcoholation and condensation reaction, and do not cause problems about odor or corrosion. In view of these properties, such room-temperature-curable organopolysiloxane compositions find use as sealing and coating agents in the manufacture of electrical and electronic devices.

[0003] Processes of sealing and coating employed in the manufacture of modern electrical and electronic devices require the use of room-temperature-curable organopolysiloxane compositions which are characterized by the most optimal speed of curing. However, it is not easy to adjust the aforementioned speed of curing while maintaining desired storage stability.

[0004] It is an object of the present invention to provide a room-temperature-curable organopolysiloxane composition that possesses excellent storage stability in a moisture-free environment and provides easy control of curing speed.

Disclosure of Invention

[0005] The room-temperature-curable organopolysiloxane composition of the invention comprises:

(A) 100 parts by mass of a diorganopolysiloxane that has a viscosity of 100 to 1,000,000 mPa·s at 25°C and molecular terminal groups selected from the group consisting of

alkoxysilyl groups, hydrogen atoms, hydroxyl groups, and substituted or unsubstituted univalent hydrocarbon groups but where at least 50 mole % of the terminal groups are said alkoxysilyl groups, which are represented by the following formula:

$$-X-SiR^1_a(OR^2)_{(3-a)}$$

(wherein R^1 represents substituted or unsubstituted univalent hydrocarbon groups, R^2 represents alkyl groups, X represents oxygen atoms or alkylene groups, and "a" is 0 or 1);

(B) 0.1 to 20 parts by mass of an organotrimethoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrimethoxysilane:

$$R^3Si(OCH_3)_3$$

(wherein R^3 represents substituted or unsubstituted univalent hydrocarbon groups);

(C) 0.1 to 20 parts by mass of an organotrialkoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrialkoxysilane:

$$R^4Si(OR^5)_3$$

(wherein R^4 represents substituted or unsubstituted univalent hydrocarbon groups, and R^5 represents alkyl groups with two or more carbon atoms); and

(D) 0.1 to 10 parts by mass of a titanium chelate catalyst.

[0006] In the above composition, the ratio of the content of component (B) to the content of component (C) in terms of mass units may range from (1 : 20) to (5 : 1).

[0007] The composition may further comprise (E) a silica-based filler used in an amount of 1 to 50 parts by mass per 100 parts by mass of component (A).

[0008] The composition may further comprise (F) an adhesion promoter used in an amount of 0.01 to 10 parts by mass per 100 parts by mass of component (A).

Effects of Invention

[0009] The room-temperature-curable organopolysiloxane composition of the invention is efficient in that it possesses excellent storage stability in a moisture-free environment and provides easy control of curing speed.

Detailed Description of the Invention

[0010] The room-temperature-curable organopolysiloxane composition of the present invention will now be described in more details.

[0011] Diorganopolysiloxane (A) is a component that has molecular terminal groups selected from the group consisting of alkoxysilyl groups, hydrogen atoms, hydroxyl

groups, and substituted or unsubstituted univalent hydrocarbon groups, wherein the alkoxysilyl group is represented by the following formula:

$$-X-SiR^1_a(OR^2)_{(3-a)}.$$

In the above formula of the alkoxysilyl groups, R^1 represents substituted or unsubstituted univalent hydrocarbon groups that may be specifically exemplified by methyl, ethyl, propyl, butyl, hexyl, octyl, decyl, octadecyl, or similar alkyl groups; cyclopentyl, cyclohexyl, or similar cycloalkyl groups; vinyl, allyl, butenyl, hexenyl, or similar alkenyl groups; phenyl, tolyl, naphthyl, or similar aryl groups; benzyl, phenethyl, or similar aralkyl groups; chloromethyl, 3-chloropropyl, 3,3,3-trifluoropropyl, or similar halogenated alkyl groups; chlorophenyl, chlorotolyl, or similar halogenated aryl groups. Most preferable are alkyl groups, aryl groups, and especially methyl and phenyl groups. R^2 represents alkyl groups, such as methyl, ethyl, propyl, and butyl groups. Most preferable are methyl and ethyl groups. In the above formula, X represents oxygen atoms or alkylene groups. These alkylene groups can be exemplified by ethylene, propylene, methylethylene, and butylene groups. Most preferable are ethylene and propylene groups. X preferably represent oxygen atoms. In the formula, "a" is 0 or 1, of which 0 is preferable.

[0012] The aforementioned alkoxysilyl groups can be specifically exemplified by trimethoxysiloxy groups, triethoxysiloxy groups, dimethoxyethoxysiloxy groups, methoxydiethoxysiloxy groups, triisopropoxysiloxy groups, tri(methoxyethoxy)siloxy groups, or similar trialkoxysiloxy groups; trimethoxysilylethyl groups, trimethoxysilylpropyl groups, triethoxysilylethyl groups, or similar trialkoxysilylalkyl groups, of which trimethoxysiloxy and trimethoxysilylethyl groups are preferable.

[0013] The aforementioned univalent hydrocarbon groups of the terminal groups may be specifically exemplified by the same univalent hydrocarbon groups as those defined above for R^1. Most preferable are alkyl, alkenyl, and aryl groups, especially methyl, vinyl, allyl, and phenyl groups.

[0014] Molecular terminal groups of component (A) are selected from the group consisting of alkoxysilyl groups, hydrogen atoms, hydroxyl groups, and substituted or unsubstituted univalent hydrocarbon groups where at least 50 mole %, preferably 60 mole %, and most preferably at least 70 mole % of the terminal groups are the aforementioned alkoxysilyl groups, which are represented by the following formula:

$$-X-SiR^1_a(OR^2)_{(3-a)}.$$

If the ratio of the content of alkoxysilyl groups to the total amount of molecular terminal groups is below the recommended lower limit, the obtained composition will show a tendency to incomplete curing.

[0015] The silicon-bonded groups contained in the molecule of component (A) may be exemplified by the same univalent hydrocarbon groups that were defined above for R^1. Most preferable are alkyl and aryl groups, especially methyl and phenyl groups.

[0016] Component (A) has an essentially linear molecular structure, which, however, may be to some extent branched. Viscosity of component (A) at 25°C ranges from 100 to 1,000,000 mPa·s, and preferably from 100 to 100,000 mPa·s. If the viscosity of component (A) is below the recommended lower limit, this will impair mechanical properties of cured products of the composition. If, on the other hand, viscosity exceeds the recommended upper limit, this will impair handling workability of the composition and will make the composition unsuitable for use as a sealing or coating agent.

[0017] Component (B) is an organotrimethoxysilane of the formula given below or a product of partial hydrolyzation and condensation of the aforementioned organotrimethoxysilane:

$$R^3Si(OCH_3)_3$$

In this formula, R^3 represents substituted or unsubstituted univalent hydrocarbon groups which may be exemplified by the same groups that have been defined above for R^1. Most preferable are alkyl, alkenyl, and aryl groups, especially methyl, vinyl, allyl, and phenyl groups.

[0018] Component (B) in the form of an organotrimethoxysilane may be represented by methyltrimethoxysilane, ethyltrimethoxysilane, propyltrimethoxysilane, vinyltrimethoxysilane, phenyltrimethoxysilane, or a combination of two or more of the aforementioned compounds.

[0019] The amount in which component (B) is added to the composition ranges from 0.1 to 20 parts by mass, preferably 0.1 to 15 parts by mass, per 100 parts by mass of component (A). If the content of component (B) is below the recommended lower limit, the composition will either be insufficiently cured or have low stability in storage. If, on the other hand, the content of component (B) exceeds the recommended upper limit, this will either delay curing, or will impair mechanical properties of cured products of the composition.

[0020] Component (C) is an organotrialkoxysilane of the formula given below or a product of partial hydrolyzation and condensation of the aforementioned organotrialkoxysilane:

$$R^4Si(OR^5)_3$$

In the above formula, R^4 represents substituted or unsubstituted univalent hydrocarbon groups, which may be exemplified by the same univalent hydrocarbon groups that were defined for R^1 above. Preferable are alkyl, alkenyl, and allyl groups, especially methyl, vinyl, allyl, or phenyl groups. In the above formula, R^5 represents alkyl groups with two or more carbon atoms. Specific examples are ethyl, propyl, butyl, hexyl, or similar alkyl having 2 to 6 carbon atoms. Most preferable are ethyl and propyl groups.

[0021] Component (C) as an organotrialkoxysilane may be specifically exemplified by methyltriethoxysilanes, ethyltriethoxysilanes, propyltriethoxysilanes, vinyltriethoxysilanes, phenyltriethoxysilanes, or mixtures of two or more of the above compounds.

[0022] Component (C) is added to the composition in an amount of 0.1 to 20 parts by mass, preferably 0.1 to 15 parts by mass per 100 parts by mass of component (A). If the added amount of component (C) is below the recommended lower limit, the composition will either be insufficiently cured or have low stability in storage. If, on the other hand, the content of component (C) exceeds the recommended upper limit, this will either delay curing, or will impair mechanical properties of cured products of the composition.

[0023] There are no special limitations with regard to the ratio in which components (B) and (C) can be used in the composition. However, in order to maintain storage stability of the composition while easily adjusting the speed of curing, it is recommended that in terms of mass units the ratio of the content of component (B) to the content of component (C) is in the range of (1 : 20) to (5 : 1), and preferable in the range of (1 : 10) to (2 : 1).

[0024] Component (D) is a titanium chelate catalyst used to accelerate curing of the composition. Specific examples of component (D) are the following: titanium dimethoxy-bis(methyl acetoacetate), titanium diisopropoxy bis(acetyl acetonate), titanium diisopropoxy bis(ethyl acetoacetate), titanium diisopropoxy bis(methyl acetoacetate), and titanium dibutoxy bis(ethyl acetoacetate).

[0025] Component (D) can be added to the composition in an amount of 0.1 to 10 parts by mass, preferably 0.3 to 6 parts by mass per 100 parts by mass of component (A). If component (D) is added in an amount less than the recommended lower limit, it will be impossible to accelerate curing of the composition to a sufficient degree. If, on the other

hand, component (D) is added in an amount exceeding the recommended upper limit, this will impair storage stability of the composition.

[0026] In order to improve flowability of the composition or the mechanical properties of a cured product, the composition may be further combined with a silica-based filler (E). The silica-based filler may be exemplified by fumed silica, fused silica, precipitated silica, quartz, or the aforementioned substances surface treated with silane compounds, silazanes compounds, or siloxanes of low degree of polymerization. Most preferable for use as component (E) is fumed silica, especially one having BET specific area equal to or greater than 50 m^2/g. Although there are no special restrictions with regard to amounts in which component (E) is added, it is recommended to add component (E) in an amount of 1 to 50 parts by mass, preferably 1 to 30 parts by mass, per 100 parts by mass of component (A).

[0027] In order to improve adhesion of a cured body of the composition to various substrates upon curing, the composition may be combined with an adhesion promoter (F). Such component (F) can be exemplified by 3-aminopropyl trimethoxysilane, 3-(2-aminoethyl) aminopropyl trimethoxysilane, or a similar amino-containing organoalkoxysilane; 3-glycidoxypropyl trimethoxysilane, or a similar epoxy-containing organoalkoxysilane; 3-mercaptopropyl trimethoxysilane, or a similar mercapto-containing organoalkoxysilane; or a reactive mixture of amino-containing organoalkoxysilane and epoxy-containing organoalkoxysilane. Most preferable are amino-containing organoalkoxysilane and a reactive mixture of amino-containing organoalkoxysilane with epoxy-containing organoalkoxysilane. Although there are no special restrictions with regard to amounts in which component (F) is added, it is recommended to add component (F) in an amount of 0.01 to 10 parts by mass, preferably 0.05 to 100 parts by mass, per 100 parts by mass of component (A).

[0028] Within the limits not contradicting the objects of the invention, the composition may be further combined with other additives such as calcium carbonate, titanium dioxide, diatomaceous earth, alumina, magnesia, zinc oxide, colloidal calcium carbonate, carbon black, or similar fillers; the aforementioned fillers surface-treated with silane compositions, silazanes compositions, or low-degree-polymerization siloxane; as well as organic solvents, anti-corrosive agents, flame retarders, heat-resistant agents, plasticizers, thixotropicity-imparting agents, pigments, etc.

[0029] There are no special restrictions with regard to a method of composition preparation, and the composition can be prepared by mixing components (A) to (D), if

necessary, with other required components. However, in case of storing the composition as a one-part RTV, mixing with component (D) and storage after mixing with component (D) have to be carried out under moisture-free conditions. In case the composition is stored as a two-part RTV, components (A) and (D) should be stored separately.

Examples

[0030] The room-temperature-curable organopolysiloxane composition of the invention will now be described in more details with reference to practical and comparative examples. The following methods were used for evaluating speed of curing and storage stability of the room-temperature-curable organopolysiloxane composition.

[Speed of Curing]

[0031] After the preparation, the room-temperature-curable organopolysiloxane composition was stored for 7 days at 25°C under moisture-insolated conditions. Following this, the composition was applied as a 2 mm-thick layer onto a glass plate, retained at 25°C and 50% relative humidity, the surface of the coating was lightly touched with a finger tip, and a Tack Free Time (TFT) was measured.

[Storage Stability]

[0032] After the preparation, the room-temperature-curable organopolysiloxane composition was stored for 28 days in an oven under moisture-insolated conditions at 50°C. The composition was then cooled to room temperature, the TFT was measured as described above, and, based on this, judgment was made on change in the speed of curing.

[Practical Examples 1 to 5, Comparative Examples 1 to 4]

[0033] In order to provide room-temperature-curable organopolysiloxane compositions having TFT in the range of 20 to 30 min., the compositions shown in Table 1 were prepared by mixing the following components under moisture-free conditions:

component (a): a dimethylpolysiloxane having at 25°C a viscosity of 2,000 mPa·s, 85 mole% of the molecular terminal groups being trimethoxysiloxy groups of the following formula:

$$-O-Si(OCH_3)_3,$$

the remaining groups being methyl groups;

component (b): $CH_3Si(OCH_3)_3$;

component (c): $CH_3Si(OC_2H_5)_3$;

component (d): titanium diisopropoxy-bis(ethylacetoacetate);

component (e): fumed silica having BET specific area of 200 m^2/g and hydrophobically surface treated with hexamethyldisilazane; and

component (f): a reactive mixture obtained after mixing 3-aminopropyl-trimethoxysilane and 3-glycidoxypropyl-trimethoxysilane in a mole ratio of 1:2 and causing the mixed components to react for 4 weeks at 25°C.

[0034] The obtained room-temperature-curable organopolysiloxane compositions were evaluated with regard to speed of curing and storage stability. The results are shown in Table 1.

[0035] [Table 1]

Examples		Practical Examples					Comparative Examples			
Components		1	2	3	4	5	1	2	3	4
Composition (parts by mass)	(a)	100	100	100	100	100	100	100	100	100
	(b)	2	1	1	0.5	0.75	2	-	-	-
	(c)	3	3	2	2	4	-	2	3	4
	(d)	2	4	2	2	3	2	2	2	2
	(e)	4	4	4	4	4	4	4	4	4
	(f)	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
TFT (min.)	after 7 days at 25°C	21	21	21	27	23	7	22	35	43
	after 28 days at 25°C	29	24	28	29	28	9	39	49	58

[0036] As can be seen from Table 1, if the composition does not contain component (C), it is difficult to provide the desired TFT within 20 to 30 min. (see Comparative Example 1). If the composition does not include component (B), this will impair storage stability (see Comparative Examples 2 to 4). Comparison of Practical Examples 1 and 2 with Comparative Example 3, of Practical Examples 3 and 4 with Comparative Example 2, and of Practical Example 5 with Comparative Example 4 shows that inclusion of components (B) and (C) makes it possible to provide TFT in the range of 20 to 30 min. and at the same time to impart good storage stability to the composition.

Industrial Applicability

[0037] Since the room-temperature-curable organopolysiloxane composition of the invention possesses excellent storage stability in a moisture-free environment and provides

easy control of curing speed, it is suitable for use as sealing and coating agents in the manufacture of electrical and electronic devices.

CLAIMS

1. A room-temperature-curable organopolysiloxane composition comprising:

(A) 100 parts by mass of a diorganopolysiloxane that has a viscosity of 100 to 1,000,000 mPa·s at 25°C and molecular terminal groups selected from the group consisting of alkoxysilyl groups, hydrogen atoms, hydroxyl groups, and substituted or unsubstituted univalent hydrocarbon groups but where at least 50 mole % of the terminal groups are said alkoxysilyl groups, which are represented by the following formula:

$$-X\text{-}SiR^1_a(OR^2)_{(3-a)}$$

(wherein R^1 represents substituted or unsubstituted univalent hydrocarbon groups, R^2 represents alkyl groups, X represents oxygen atoms or alkylene groups; and "a" is 0 or 1);

(B) 0.1 to 20 parts by mass of an organotrimethoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrimethoxysilane:

$$R^3Si(OCH_3)_3$$

(wherein R^3 represents substituted or unsubstituted univalent hydrocarbon groups);

(C) 0.1 to 20 parts by mass of an organotrialkoxysilane of the formula given below or a product of partial hydrolyzation and condensation of said organotrialkoxysilane:

$$R^4Si(OR^5)_3$$

(wherein R^4 represents substituted or unsubstituted univalent hydrocarbon groups, and R^5 represents alkyl groups with two or more carbon atoms); and

(D) 0.1 to 10 parts by mass of a titanium chelate catalyst.

2. The room-temperature-curable organopolysiloxane composition according to Claim 1, wherein the ratio of the content of component (B) to the content of component (C) in terms of mass units ranges from (1 : 20) to (5 : 1).

3. The room-temperature-curable organopolysiloxane composition according to Claim 1, further comprising (E) a silica-based filler used in an amount of 1 to 50 parts by mass per 100 parts by mass of component (A).

4. The room-temperature-curable organopolysiloxane composition according to Claim 1, further comprising (F) an adhesion promoter used in an amount of 0.01 to 10 parts by mass per 100 parts by mass of component (A).

INTERNATIONAL SEARCH REPORT

International application No
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A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L83/04 C08K5/09

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)
C08L C08K

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 practical, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 6 132 664 A (FREIBERG ALAN LEE [US] ET AL) 17 October 2000 (2000-10-17) column 6, lines 48-64; examples 1-4; tables 1-3	1-4
X	US 2004/116639 A1 (LIM THOMAS F [US] ET AL) 17 June 2004 (2004-06-17) claim 16; tables 2a-3b	1-4

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

☒ See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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Date of the actual completion of the international search	Date of mailing of the international search report
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 6132664 A	17-10-2000	EP 0926205 A1	30-06-1999
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