

THERMALLY AND ELECTRICALLY CONDUCTIVE AND ADHESIVE COMPOSITION FOR SEMICONDUCTOR PACKAGING

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TECHNICAL FIELD

[001] This invention concerns a composition for packaging semiconductor devices such as integrated circuits (IC) and large scale integration (LSI) devices.

10 The composition is useful for die attaching these devices onto a substrate while exhibiting suitable thermal and electrical conductivity along with good adhesion onto leads and lead frame parts of the package.

15 BACKGROUND ART

[002] With the passing and enforcement of lead-free solder legislation under WEEE (Waste Electrical and Electronic Equipment) and RoHS (Restriction of Hazardous Substances) tin-lead solder are replaced by lead-free solder,

20 thereby increasing the solder reflow temperature from about 240°C to around 260°C due to the higher melting point of lead free solder. Higher temperature in turn leads to higher stress on semiconductor packages during reflow due to higher coefficient of thermal expansion (CTE) mismatch found between various components present in a semiconductor device. The problem is further
25 compounded by the weakening of adhesion strength between the

semiconductor device and the leads or lead frame at such higher temperature due to deterioration of chemical bonding thereinbetween.

[003] These problems may lead to more frequent and serious package

5 delamination and cracks after the reflow process, and may be critical enough to result in package failure. Hence, various attempts have been made to improve upon the formulation of the adhesive composition to achieve better adhesion and electrical and/or thermal conductivity, as well as reduction of package stress during high temperature reflow process.

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[004] One well-known approach to improving adhesion is to include a high proportion of silver (argentum, Ag) as inorganic filler in the composition, typically in the form of silver flakes or powders in the range of 70% to 90% such as that disclosed in U.S. Patent No. **6,225,379** (National Starch & Chemical) and U.S.

15 Patent No. **5,908,881** (Sumitomo Bakelite). It is known that, besides its good dispersion in organic resin compositions, Ag provides superior thermal and/or electrical conductivity. At below 70%, however, the resultant composition has poor thermal conductivity, thus lowering the heat discharged rate from a semiconductor chip in operation, whereas at above 90% the resultant viscosity
20 of the composition becomes too high to be of practical use although in U.S.

Patent No. **4,595,604** (Rohm and Haas) as high as 95% content of Ag has been reported for such conductive compositions.

[005] Another aspect is to choose an appropriate epoxy resin system to constitute the packaging composition. In the aforementioned **US-5,908,881** (Sumitomo Bakelite) an epoxy resin having at least 2 epoxy groups in the molecule and in liquid form at room temperature is disclosed. This epoxy resin
5 having preferably 3 epoxy groups, together with a reactive diluent and curing agent, are designed to avoid any phenolic hydroxyl groups remaining unreacted and thus making the composition thermally insulative wherein the curing temperature is higher than its melting point. Thus, the epoxy resin system including the diluents, thixotropic or viscosity agents and curing or hardening
10 agents are also important considerations in the formulation of a desired adhesive composition.

[006] Yet another aspect of the composition is to provide for suitable coupling agents for surface to surface adhesion. In U.S. Patent No. **6,645,630**
15 (Matsushita Electric) the epoxy resin composition disclosed is based on a phosphorus compound wherein it is suggested that one or more of the following 3 types of coupling agent is added to the composition, i.e. epoxysilane, mercaptosilane and aminosilane coupling agents. The inorganic filler proposed, however, is aluminum hydroxide and not silver.

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[007] In U.S. Published Application No. **2005/0090044** (Sumitomo Bakelite), an epoxy resin of the tetramethylbisphenol F type is disclosed wherein a silane coupling agent is suggested to be added to the composition so as to improve the molding and adhesion properties. While many types of aminosilanes have

SUMMARY OF DISCLOSURE

10 [008] In our present invention, we endeavour to provide an epoxy resin composition for die attaching wherein the cured composition exhibits good thermal and electrical conductivity while at the same time provides good adhesion to lead surfaces or components of the semiconductor package, whereby delamination and cracks are avoided during reflow temperatures in the
15 packaging process.

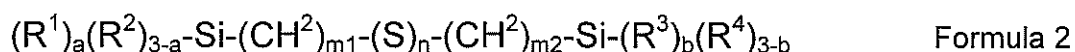
[009] The novel adhesive thermosetting epoxy resin composition would have desired properties such as high adhesion strength, low elastic modulus, long pot life, long storage life, good anti-bleed out performance, and good solder
20 cracking resistance.

[010] As a general embodiment, our heat and electrically conductive adhesive composition comprises of (A) a filler comprising substantially of silver (Ag) aggregate, in an amount of 70 to 90% by weight of the adhesive composition;
25 (B) an epoxy resin in liquid form comprising at least two epoxy groups; (C) a

curing or hardening agent; and (D) a silane coupling agent comprising at least a primary mercapto group.

[011] In one aspect of our composition, the silane coupling agent comprises

5 sulfidosilane, preferably, bis-sulfur silane. A preferred embodiment of the silane coupling agent may be generally represented by Formula 2 below:



10 wherein:

- R^1 and R^3 are each an alkoxy group with 1 to 10 carbon atoms, wherein R_1 and R_3 may or may not be the same;

- R^2 and R^4 are each an alkyl group with 1 to 10 carbon atoms, wherein R_2 and R_4 may or may not be the same;

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- a and b are integers of 1 to 3;
- n is an integer of 2 to 4; and
- m_1 and m_2 are integers of 1 to 5.

[012] A specific embodiment of the silane coupling agent is bis-

20 [triethoxysilylpropyl]tetra sulfide, wherein R^1 and R^2 are both ethoxy; R_3 and R_4 are interchangeably an ethoxy and propyl; a and b are each = 3; n = 4; such that the compound has $(C_2H_5O)_3SiC_3H_6S_4C_3H_6Si(OC_2H_5)_3$ as its molecular formula. Preferably, the amount of silane coupling agent is in the range of 0.1%

- 8% by weight of the total weights of (B) epoxy resin and (C) phenolic compound.

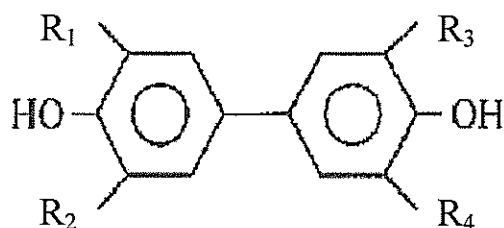
[013] In a second aspect of our composition, the filler aggregate size is preferably in a range of up to 50 μm , and most preferably below 10 μm . The silver content may be maintained at a range of between 70% - 90% of the filler used. The silver ratio is preferred to be at least 90% or more by weight of the filler content, though a combination of silver with an alloy of silver or other metals is allowable. The filler aggregate is preferably in the form of flakes which may be used together with spherical form of silver fillers.

[014] In a third aspect of the composition, the epoxy resin may comprises of at least two epoxy groups selected from epoxidized bis-phenol A, bis-phenol F, biphenol or derivatives thereof. Preferably, the epoxy resin comprises 3 epoxy groups further including tri-hydroxyl-phenyl-methane or amino-phenol. Yet another preferred embodiment of the epoxy resin comprises multi-epoxy groups further including selections from epoxidised phenol novolak, and/or cresol novolak.

[015] In a fourth aspect of our composition, the curing or hardening agent is a compound with at least two phenolic groups and is preferably provided in an amount of between 1 to 20% by weight of the epoxy resin, more preferably 3 – 15% by weight of the epoxy resin. A preferred embodiment of the curing or

hardening compound is a biphenol derivative and which may have a general formula of

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wherein:

- R₁, R₂, R₃, R₄ are either H or CH₃, and may or may not be the same.

- 10 [016] The biphenol derivative has a melting point higher than 200°C, more preferably higher than 260°C, and dispersable as powder particles in the conductive adhesive composition.

- [017] In a fifth aspect of our invention, our composition further includes
- 15 constituent (E) a catalyst including an imidazole compound having melting point above 180°C and provided in the range of 0.1% - 10% by weight of (B) epoxy resin, and which is preferably any one or a combination selected from:

- 2-phenyl-4,5-dihydroxymethylimidazole;
- 2-phenyl-4-methylimidazole;
- 20 - 2-phenyl-4-methyl-5-hydroxymethylimidazole;
- 2,4-diamine-6[2'-ethyl-4'-methylimidazole-(1')ethyl-s-triazine.

- [018] The composition according to our present invention, apart from the five constituents described above, if necessary, may further include low stress

additives, defoaming agents, diluents and leveling agents which are suitable for use in the packaging of semiconductor devices, including dispensing a paste form of said composition for die attaching processes.

5 LIST OF ACCOMPANYING DRAWINGS

[019] A better understanding of the invention may be obtained from referring to the drawings listed below which accompanies this specification, and the detailed description of specific embodiments that follows, as non-limiting and
10 exemplary illustration of specific aspects or preferred embodiments, in which:

[020] Figure 1 shows the Differential Scanning Calorimetry (DSC) curve of example of compound (D), product code no. KBE-846 (by Shin-Etsu Chemical Co., Ltd) ; and
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[021] Figure 2 shows the DSC curve of the same compound KBE-846 mixed with silver in the same proportion by weight.

20 DETAILED DESCRIPTION OF SPECIFIC EMBODIMENTS

[022] The general embodiment of the composition of our invention comprises the following 4 constituents (designated A to D) with an optional 5th catalyst compound (designated E):

- (A) a filler comprising substantially of silver (Ag) aggregate, in an amount of 70 to 90% by weight of the adhesive composition;
- (B) an epoxy resin in liquid form comprising at least two epoxy groups;
- (C) a curing or hardening agent; and
- 5 (D) a silane coupling agent comprising at least a primary mercapto group, and optionally,
- (E) an imidazole catalyst.

[023] Silver (A) is used as an inorganic filler to achieve good electrical
10 conductivity and thermal conductivity. Though a combination of silver with an alloy of silver or other metals is allowable, it is preferred that the ratio of silver be maintained at least 90% or more by weight of the filler content. In addition, ionic impurities such as sodium, chloride, and potassium ion should be kept low for its application in semiconductor field.

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[024] It is preferred that average filler size be below 10 μ m and not exceed 50 μ m. Though flake type is preferred, spherical shaped filler could be also used together with flake filler, which are easily available for semiconductor application. Below 70% of silver in weight gives less than desired electrical conductivity
20 while above 90% of silver in weight will lead to high viscosity of the composition that makes dispensability poor.

[025] The next 2 constituents of the composition, i.e. epoxy resin (B) and a curing or hardening agent (C) is a compound with at least 2 phenolic groups

(OH groups) (C) used as hardener are now described. Epoxy resin is used due to its good adhesion property, while the phenolic compound is selected as hardener as it shows the best anti-water absorption property among all cure systems with epoxy resin. As for the case of silver, the ionic impurities of epoxy resin (B) and the curing or hardening compound (C) with at least 2 phenolic groups (OH group) should also be kept low.

[026] Epoxy resin (B) that are available for use includes, but not limited to the following:

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[027] - composition with 2 epoxy groups, e.g. epoxidized bis-phenol A, bis-phenol F, biphenol or derivative thereof;

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[028] - composition with 3 epoxy groups, e.g. tri-hydroxyl-phenyl-methane and amino-phenol; and

[029] - composition with multi-epoxy groups, e.g. epoxidized phenol novolak and cresol novolak, and etc.

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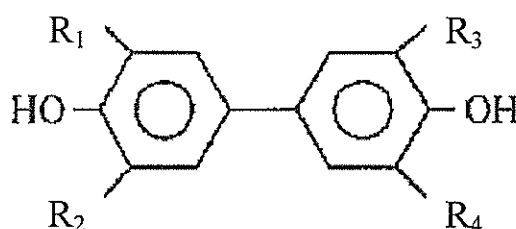
[030] It is preferred that the epoxy resin be in liquid form since an adhesive composition should be in liquid to be worked at room temperature. A reactive diluent may also be added to the adhesive composition to achieve better workability. Examples of such reactive diluents include cresol glycidylether, e.g. t-butylphenol glycidylether. Preferred diluents in this case are low molecular

weight resins with reactive functional groups whereas the solvent may be generally inert and non-reactive.

[031] In respect of constituent (C) which is used as a curing or hardening agent, we propose therefore a compound having at least 2 phenolic groups (OH groups) and which should have melting point of higher than 200°C and therefore does not melt in the presence of common epoxy resin at room temperature. It is preferred that compound (C) have a melting point of higher than 260°C, so that it does not melt during solder reflow process, which would have led to unexpected change in property.

[032] Compound (C) is preferably dispersed as powder particles in conductive adhesive composition to give good storage properties. As the reaction with epoxy happens only at surface of compound particles, it reduces the cross-link density and serves as stress absorbent at high temperature to release the stress in the cured adhesive composition. This helps to reduce the semiconductor packages stress during high temperature reflow and prevent package delamination and crack. Compound (C) with at least 2 phenolic groups (OH groups) could, therefore, be a good alternative to liquid type elastomer that is commonly used as low stress additive. Liquid type elastomer normally gives poor solubility in epoxy resin (B), which affects curability and cause poor adhesion and resin bleed out issues.

[033] Examples of compound (C) with at least 2 phenolic groups (OH groups) include, at least, the biphenol derivatives indicated as Formula 1 below:



Formula 1

[034] where R₁, R₂, R₃, R₄ are either H or CH₃. The content of biphenol derivatives, Formula (1), is within 1% to 20% by weight of the epoxy resin (B) in adhesive composition, though 3% to 15% by weight is preferred. At below 1% by weight of biphenol content does not give the desired low modulus effect while above 20% in weight will lead to too high the viscosity of the adhesive composition to be utilized.

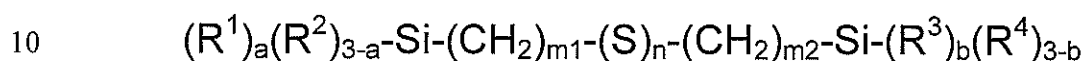
[035] In general, the high melting point hardener ensures that it remains as solid particles in adhesive composition during and after cross-linking process at elevated temperature. The solid particles in the cured adhesive composition matrix serves as stress absorber, which gives low stress property at elevated temperature and offers good solder crack resistance.

[036] At room temperature, the solid form high melting point hardener is less reactive compared to low melting point hardener, and offers longer pot life.

Similar result is found on storage life of the adhesive composition that is stored at sub-zero temperature. The high melting point hardener in the form of solid particles serves as spacer that helps to maintain both the thickness of adhesive composition and the levelness of bonding item.

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[037] In respect of constituent (D) it is used to improve adhesion of adhesive composition to adherents. Compound (D) has a general compound represented by Formula (2) below:



Formula 2

wherein

15 R^1, R^3 : Alkoxy group with 1 to 10 carbon atoms, where R_1 and R_3 may/may not be the same;

R^2, R^4 : Alkyl group with 1 to 10 carbon atoms, where R_2 and R_4 may/may not be the same;

a, b : Integers of 1 to 3;

20 n : Integers of 2 to 4; and

m_1, m_2 : Integers of 1 to 5.

[038] As generally known in the art, silane coupling agents with amino functional group or silane coupling agent with mercapto functional group can be used to enhance adhesion. Both silane coupling agents with amino functional

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group and mercapto functional group, however, show slight adhesion improvement only when relatively high amount is applied and this high amount will normally give the adverse effect on pot life as it accelerates reaction at room temperature.

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[039] Since compound (D) does not give significant reaction with epoxy groups, compounds with phenolic groups (OH groups) and silver at room temperature, it can, therefore, be loaded at high amount into the adhesive composition to give significant improvement on adhesion, and yet maintaining a stable storage properties. Moreover, besides offering better adhesion performance than other silane coupling agent, it helps to prevent significant drop in adhesion strength after moisture treatment that contributes to high reliability performance.

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[040] As the structure of $(S)_n$ in compound (D) forms chemical reaction with the surface of metal lead-frame, as well as the surface of silver filler, it therefore improves not only the adhesion strength to lead-frame but also the cohesive strength of cured adhesive composition that is achieved by improving the chemical bonding between silver filler and resins.

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[041] Figure 1 shows the Differential Scanning Calorimetry (DSC) curve of an example of compound (D), in the form of Product Code no. KBE-846 (Shin-Etsu Chemical Co., Ltd, where both R_1 and R_2 indicated in Formula 2 are ethoxy, both a & b are 3, n is 4 and both m_1 & m_2 are 3).

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[042] Figure 2 shows the DSC curve of the same compound (D) in the form of Product Code no. KBE-846 that is mixed with same % by weight of silver. In the DSC test, the sample amount is about 30mg and the test condition is 10°C/min from room temperature. The results show that compound (D) reacts with silver,
5 and through this reaction, the effects of this invention could be realized.

[043] For above effects, n indicated in Formula 2 is to be limited between 2 and 4, with $n = 4$ is preferred. In case that where n is less than 2 or more than 4, the effect on adhesion improvement is not expected. Similar to standard silane
10 coupling agents, there could be 1 to 3 alkoxy groups bonded to each of both the end Si atoms. The alkoxy group is preferred to be methoxy, ethoxy or butoxy group to ensure good reactivity. Alkoxy with more than 11 carbon atoms could not be used due to poor reactivity. Either methoxy or ethoxy is highly preferred in this case and 2 or 3 of these alkoxy groups is preferred to be
15 bonded to the Si atoms. m_1 & m_2 are integer numbers from 1 to 5, where 3 is preferred.

[044] The content of compound (D) represented by Formula 2 can be in the range of 0.1% to 8% in weight based on total weight of epoxy resin (B) and
20 compound with at least 2 phenolic groups (OH groups) (C). Below 0.1% in weight of compound (D) does not give significant improvement on adhesion, while above 8% in weight of compound (D) causes poor reactivity and voids. Formation of voids is due to alcohol formed through hydrolysis of alkoxy silane groups.

[045] The fifth constituent, i.e. imidazole compound (E) is utilized as catalyst to the reaction and curing process between the epoxy resin (B) and compound with at least 2 phenolic groups (OH groups) (C). The imidazole compound (E) is preferred to have melting point of higher than 180°C to give long and stable storage life of the adhesive composition. It includes, but not limited to, compositions with imidazole group e.g. 2-phenyl-4,5-dihydroxymethylimidazole, 2-phenyl-4-methylimidazole, 2-phenyl-4-methyl-5-hydroxymethylimidazole, 2,4-diamine-6[2'-ethyl-4'-methylimidazolyl-(1')]ethyl-s-triazine. The content of imidazole compound (E) can be in the range of 0.1% to 10% by weight based on total weight of epoxy resin (B) in adhesive composition. Below 0.1% in weight of compound (E) will take long time for complete curing, while above 10% in weight will lead to too high the viscosity of the adhesive composition to be utilized.

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[046] Apart from the five constituents described above, the adhesive composition of this invention, if necessary, can contain additives such as low stress additives, defoaming agents, diluents and leveling agents.

20 [047] The process of producing the adhesive composition of this invention comprises, but not limited to, mixing of all constituents of adhesive composition together followed by kneading of the mixture using a three rolls mixer or similar equipments. Vacuum defoaming machine is then used to remove the bubbles in the adhesive composition.

[048] The adhesive composition of this invention is applicable to packaging conventional semiconductor devices assembly process. This typically includes, dispensing of adhesive composition followed by mounting of chips onto designated positions of lead-frame or substrate using a die-bonder machine.

- 5 The adhesive composition is then cured with heat. Wire-bonding process is performed to form interconnects and the chips are then encapsulated using epoxy molding compound through transfer molding process.

10 EXAMPLES

[Examples 1 to 4]

- [049] Flake silver with average size of 1 to 30 μ m (hereunder "silver") as silver
- 15 (A); diglycidylether of bisphenol A (epoxy equivalent = 180, liquid at room temperature, hereunder "bis-A epoxy"), synthesized by reaction between bisphenol A and epichlorohydrin, as epoxy resin (B); cresol glycidylether (epoxy equivalent = 185, hereunder "CGE"); biphenol (OH equivalent = 180, melting point = 260°C, hereunder "phenol A") as compound with at least 2 phenolic
- 20 groups (OH groups) (C) that indicated as Formula 1; KBE-846 (Shin-Etsu Chemical Co., Ltd Made, bis(triethoxysilylpropyl) tetrasulfide, hereunder "coupling agent A") as compound (D) that indicated as Formula 2; 2P4MHZ (Shikoku Chemicals Corporation Made, hereunder "2P4MHZ", melting point=192°C), as Imidazole compound (E); and Dicyandiamide as hardener; are

compounded as indicated in table 1 where the adhesive compositions were prepared using kneading machine that consists of 3 rollers. After the kneading process, the adhesive compositions were de-foamed in vacuum chamber at 2mmHg for 30minutes. Properties of the adhesive compositions were then
5 evaluated with test methods as describe below.

[Comparative example 1 to 4]

10 [050] In comparative example 2, KBM-573 (Shin-Etsu Chemical Co., Ltd Made, N-phenyl-γ-aminopropyltrimetoxysilane, hereunder “coupling agent B”) as coupling agent with amino group was used and in comparative example 3, KBM-803P (Shin-Etsu Chemical Co., Ltd Made, 3-merkaptopropyl-tri-methoxy silane, hereunder “coupling agent C”) as coupling agent with merkapto group
15 was used. In comparative example 4 phenol novolak (OH equivalent = 104, melting point = 80°C to 90°C, hereunder “phenol B”) was used.

Test method

20 [051] Viscosity: viscosity measurement is done using Brookfield viscometer (5°corn, at 25°C, at 5.0 rpm) for adhesive compositions as indicated in table 1, at both 0 hour and 48 hours of staging at room temperature. Adhesive composition with viscosity that falls in the range of 6000 mPa·s to 12000 mPa·s,

and shows change in viscosity between 0 hour to 48 hours of less than 20% is considered pass. The unit of viscosity is mPa·s.

[052] Die shear strength: test units for die shear test are prepared by mounting
 5 6x6 mm silicon chips on i) Cu lead-frame, ii) Cu lead-frame with Ag plating, iii)
 Ni-Pd plating lead-frame with Au flash, using adhesive compositions as
 indicated in Table 1, and cured at 175°C/30 min. Die shear strength
 measurement is performed at 260°C using auto die shear tester. Value of more
 than 30N/chip at 260°C is considered pass. The unit of die shear strength is
 10 N/chip.

[053] Volume resistivity: test samples with dimension of 4 x 50 x 0.04 mm are
 prepared, using adhesive compositions as indicated in table 1, by curing at
 175°C/30 min. The resistance over 40 mm of the sample length is measured
 15 and the volume resistivity is then calculated. Volume resistivity value of below
 $1 \times 10^{-3} \Omega \cdot \text{cm}$ is considered pass. The unit of volume resistivity is $\Omega \cdot \text{cm}$.

[054] Storage modulus: test samples with dimension of 4x50x0.1mm are
 prepared, using adhesive compositions as indicated table 1, by curing at
 20 175°C/30min. The storage modulus is measured using Dynamic Mechanical
 Analyzer, DMA by heating from 25°C to 300°C at the rate of 10°C/min. Storage
 modulus value at 250°C of below 450 MPa is considered pass. The unit of
 storage modulus is MPa.

[055] Void: test units for void test are prepared by mounting 6 x 6 mm glass chips on Cu lead-frame using adhesive compositions as indicated in Table 1, and cured at 175°C/30 min. Observation for void is then done under microscope. Adhesive composition that shows void area of less than 10% is considered pass.

5 The unit of void area is represented as % of total area.

[056] Reliability: IC package used for reliability test is built by mounting silicon chips onto lead-frame using adhesive compositions as indicated in Table 1, which is cured at 175°C/30 min, and molding it with epoxy molding compound

10 (EME-G700H, Sumitomo Bakelite Ltd.). The packages will then be treated under condition of (moisture sensitivity level, MSL 2A accelerate) 60°C/60RH% for 120 hours and subjected to re-flow process of 260°C, 10sec, 3 times. SAT (T-SAM) is used to check delamination of the packages. Delamination area of less than 10% is considered pass. The unit of delamination area is represented
15 as % of total area.

[057] Package : QFP (14x20x2.0mm)

Lead-frame : 1.) Ni-Pd plating lead-frame with Au flash
2.) Ag ring plating lead-frame

20 Chip size : 6x6mm

Cure condition : 175°C, 30min in oven

[058] The results of the above-mentioned tests are shown in Table 1 below:

Adhesive Compositions and Physical Properties and Reliability Results

			Example				Comparative Example			
			1	2	3	4	1	2	3	4
Silver			75	75	75	75	75	75	75	75
Bis-A epoxy			16.13	12.73	14.89	14.11	15.08	14.89	14.89	14.89
CGE			6.91	5.45	6.38	6.05	6.46	6.38	6.38	6.38
Phenol A			0.32	2.42	2.13	2.02	2.16	2.13	2.13	
Phenol B										2.13
2P4MHZ			1.15	0.91	1.06	1.01	1.08	1.06	1.06	1.06
Dicyandiamide			0.23	0.18	0.21	0.2	0.22	0.21	0.21	0.21
Coupling agent A			0.35	0.32	0.10	1.20				0.27
Coupling agent B								0.32		
Coupling agent C									0.32	
Viscosity										
Initial	Initial	mPa·s	6214	13753	11350	10190	11516	9693	9279	7871
	After 48hr @ 25°C	mPa·s	6131	13753	11350	10356	11599	9362	9859	8579
	% change	%	-1.3	3.5	0	1.6	0.7	-3.4	6.3	9
Die shear Strength @ 260°C	After cure / Ag ring	N/chip	71	63	55	64	59	48	59	34
	After cure / Ag spot	N/chip	57	82	49	58	26	10	9	42
	After cure / Ni-Pd Au	N/chip	80	82	67	75	29	23	24	61
Volume resistivity		$\Omega \cdot \text{cm}$	5×10^{-4}	8×10^{-4}	4×10^{-4}	6×10^{-4}	2×10^{-4}	3×10^{-4}	3×10^{-4}	3×10^{-4}
Storage Modulus		MPa	480	310	350	410	350	390	460	530
Void		%	<10	<10	<10	<10	<10	<10	<10	<10
Reliability (MSL 2A accelerate) Delamination area on (Ni-Pd Au plating lead-frame)		%	<10	<10	<10	<10	>50	>50	>50	<10
Reliability (MSL 2A accelerate) Delamination area on (Ag ring plating lead-frame)		%	<10	<10	<10	<10	<10	<10	<10	>50
Overall performance			OK	OK	OK	OK	NG	NG	NG	NG

Comparing Examples 3 and 4 with Comparative Examples 1, 2 and 3

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[059] Compound (D) (hereunder Coupling agent A) gives significant adhesion strength improvement to an adhesive composition, particularly on Ag and Ni-Pd (Au) surfaces. In addition, it shows more superior performance in terms of adhesion improvement than silane coupling agent B and C. This can be

10 explained by chemical reaction that is formed between (S)_n in compound (D) and

metal lead-frame surface, as well as the silver particle surfaces, which help to improve both adhesive and cohesive strength of the cured adhesive compositions.

Comparing Example 1, 2 to Comparative Example 4

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[060] Bisphenol derivatives indicated as Formula (1) (hereunder Phenol A) is effective in reducing the storage modulus of adhesive composition. In addition, it gives higher adhesion strength on Ag ring plating lead-frame and therefore better MSL performance when used together with compound (D) compared to
10 phenol novolak (hereunder Phenol B) with lower melting point.

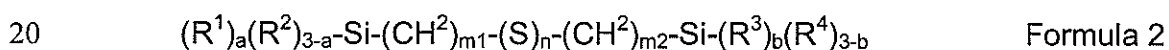
CLAIMS

We claim:

- 5 1. A heat and electrically conductive adhesive composition comprising:
- (A) a filler comprising substantially of silver (Ag) aggregate, in an amount 70 to 90 % by weight of the adhesive composition;
- (B) an epoxy resin in liquid form comprising at least two epoxy groups;
- 10 (C) a curing or hardening agent comprising at least two phenolic groups; and
- (D) a silane coupling agent comprising sulfidosilane.

- 15 2. A composition according to claim 1 wherein the sulfidosilane is bis-sulfur silane.

3. A composition according to claim 2 wherein the bis-sulfur silane has a general formula of



wherein:

- R^1 and R^3 are each an alkoxy group with 1 to 10 carbon atoms, wherein R_1 and R_3 may or may not be the same;

- R^2 and R^4 are each an alkyl group with 1 to 10 carbon atoms, wherein R_2 and R_4 may or may not be the same;

- a and b are integers of 1 to 3;

- n is an integer of 2 to 4; and

5 - m_1 and m_2 are integers of 1 to 5.

4. A composition according to claim 3 wherein the silane coupling agent is bis-[triethoxysilylpropyl]tetra sulfide, wherein

- R^1 and R^2 are both ethoxy;

10 - R_3 and R_4 are interchangeably an ethoxy and propyl;

- a and b are each = 3;

- n = 4;

such that the compound has $(C_2H_5O)_3SiC_3H_6S_4C_3H_6Si(OC_2H_5)_3$ as its molecular formula.

15

5. A composition according to claim 1 wherein the amount of silane coupling agent is in the range of 0.1% - 8% by weight of the total weight of (B) epoxy resin and (C) phenolic compound.

20

6. A composition according to claim 1 wherein the filler aggregate size is in a range of up to 50 μm .

7. A composition according to claim 6 wherein the filler aggregate size is below 10 μm .

25

8. A composition according to claim 1 wherein the silver content is maintained at a range of between 70% - 90% of the filler used and wherein said silver is in the form of an alloy of silver or other metals.

30

9. A composition according to claim 6 wherein the filler aggregate is

in the form of flakes.

10. A composition according to claim 9 wherein the flakes are used together with spherical form of fillers.

5

11. A composition according to claim 1 wherein the epoxy resin comprising at least two epoxy groups selected from epoxidized bis-phenol A, bis-phenol F, biphenol or derivatives thereof.

10

12. A composition according to claim 11 wherein the epoxy resin comprises 3 epoxy groups further including tri-hydroxyl-phenyl-methane or amino-phenol.

15

13. A composition according to claim 12 wherein the epoxy resin comprises multi-epoxy groups further including selections from epoxidised phenol novolak, and/or cresol novolak.

20

14. A composition according to claim 1 wherein the curing or hardening compound is provided in an amount of between 1 to 20% by weight of the epoxy resin.

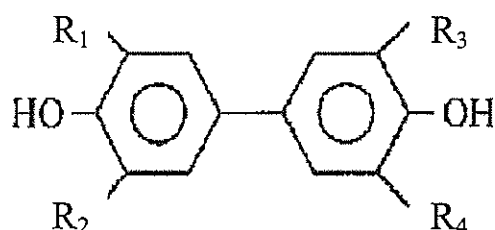
25

15. A composition according to claim 14 wherein the curing or hardening compound is provided in an amount between 3 – 15% by weight of the epoxy resin.

16. A composition according to claim 1 wherein the curing or hardening compound is a biphenol derivative.

30

17. A composition according to claim 16 wherein the biphenol derivative has a general formula of



10 wherein

- R₁, R₂, R₃, R₄ are either H or CH₃, and may or may not be the same.

18. A composition according to claim 17 wherein the biphenol derivative has a melting point higher than 200°C.

15 19. A composition according to claim 18 wherein the biphenol derivative has a melting point higher than 260°C.

20 20. A composition according to claim 16 wherein the biphenol derivative is dispersed as powder particles in the conductive adhesive composition.

21. A composition according to claim 1 further comprising:

(E) a catalyst including an imizadole compound.

25 22. A composition according to Claim 21 wherein the imizadole compound is any one or a combination selected from

- 2-phenyl-4,5-hihydroxymethylimidazole;
- 2-phenyl-4-methlyimidazole;

- 2-phenyl-4-methyl-5-hydroxymethylimidazole;
- 2,4-diamine-6[2'-ethyl-4'-methylimidazole-(1')ethyl-s-triazine.

5 23. A composition according to claim 21 wherein the imidazole compound's melting point is above 180°C.

10 24. A composition according to claim 21 wherein the imidazole compound is provided in the range of 0.1%-10% by weight of (B) epoxy resin.

25. A composition according to any of claims 1 – 24 further including, low stress additives, defoaming agents, diluents and leveling agents.

15 26. A composition according to claim 25 for use in the packaging of semiconductor devices, including dispensing a paste form of said composition, for die attaching processes.

20 27. A semiconductor devices packaged with a composition according to any one of claims 1 – 25.

28. An electronic appliance including a semiconductor device packaged with a composition according to any one of claims 1 – 25.
