

EPOXY RESIN COMPOSITION

FIELD OF THE INVENTION

The present invention relates to an epoxy resin composition used for sealing of semiconductor devices, which is an environmental friendly material ("green plastic").

BACKGROUND

Electronic parts such as diodes, transistors, integrated circuits and the like have been sealed with a epoxy resin composition, modified by the additives. These additives include flame retardants, which have commonly been included antimony, boron, phosphorus or bromine compounds. Such compounds are not "environmentally friendly".

OBJECT OF THE INVENTION

The object of this invention is to provide an environmental friendly epoxy resin composition, which shows high thermal conductivity, and good solder cracking resistance, or to at least provide the public with a useful choice.

SUMMARY OF THE INVENTION

According to the invention there is provided an epoxy resin composition used for sealing of semiconductor devices, comprising:

- (A) an epoxy resin of the formula II,
- (B) an epoxy resin curing agent,
- (C) a mixture of inorganic aluminum oxide and silicon oxide constituting 69.5-95% by weight of the total epoxy resin composition,
- (D) a curing accelerator

The naphthalene tetrafunctional type resin of the formula II is preferably present in an amount of 5 to 30% by weight of the total epoxy resin composition. It can also be used with a resin of the formula I. The epoxy resin curing agent is preferably a compound of formula III, IV or V, or a mixture thereof (herein identified as a phenolic resin curing agent of the invention), more preferably a compound of IV or V or a mixture thereof.

The curing accelerator is present in an amount preferably of 0.1-1.0 wt % based on the total epoxy resin composition, said epoxy resins (A) preferably being used in an amount of 0.8 to 1.25 epoxy equivalents per hydroxyl group equivalent of said phenolic resin curing agents (B).

The present series of environmental friendly epoxy resin composition consists of some candidates with very high heat dissipation capability as well as soldering stress resistance at 260°C as compared with conventional epoxy resin compositions used for the same purpose.

DETAILED DESCRIPTION OF THE INVENTION

In most cases polymer resin initiate or propagate fires because, being organic compounds, they decompose to volatile combustible products and release flammable vapors when they are exposed to heat. To fulfil legal requirements, flame retardant agents need to be added into the epoxy resin composition. The flame retardant performance for present environmental friendly epoxy resin composition is controlled by the type, content of ceramic fillers, type of epoxy resin and epoxy curing agent. In order to obtain maximum flame retardant ability without adding any bromine, antimony, boron, phosphorus or their compounds, alumina (thermal conductivity $\sim 30\text{W/mK}$) is selected as the main filler instead of the conventional fused silica (thermal conductivity $\sim 1.3\text{W/mK}$).

The crucial role of the alumina filler in the present epoxy resin composition is as a flame retardant. Without being bound by any theory we believe the microscopic presence of water on the surface of alumina hydrolyses the filler to yield aluminum hydroxide and during the heating or burning process, the metal hydroxide undergoes an endothermic reaction to shed its moisture content and revert to alumina. By so doing, the latent heat provides the essential flame retardancy characteristics.

It is further believed, again without being bound by theory, that during the flaming of the organic resin, hydroxyl radicals attached to the alumina filler, form aluminium hydroxide [$\text{Al}_2\text{O}_3 + 6\text{OH}^- \rightarrow 2\text{Al}(\text{OH})_3 + 3\text{O}^{2-}$], ultimately it performs as a flame retardant agent.

Using an effective amount of alumina the present epoxy resin composition does not require conventional flame retardant agent such as brominated epoxy resin, hexa-bromo antimony trioxide or pentaoxide. The outcome is an environmental friendly epoxy resin composition free of bromine, antimony, boron, phosphorus and their compounds. Among other advantages, this may render the expensive alumina filled epoxy resin composition recyclable and possess high thermal conductivity.

Epoxy resin composition, without added flame retardant, must have relatively high filler loading in order to pass the UL-94 (Underwriter Laboratory - USA)'s V-0 flammability requirement. However, different types of epoxy resin and curing agent have different flammability. This is due to a combination of the bulkiness, solidity of structure, aromatic and aliphatic carbon content.

Boundary curves can be drawn by joining the points of minimum amount of alumina filler needed in resin matrix (see graph 1) with respect to total filler content in order to work as a flame retardant.

The 1st embodiment is represented by boundary equation (II) and illustrated in graph 1 where the UL-94 V-0 requirement can be achieved:

$$Y \geq -0.0668X + 5.69 \quad \text{equation (II)}$$

Where,

Y = Ratio of alumina filler to total filler content (in wt%) is from 0 to 1.0

X = Total filler content in wt% is from 69.5% to 95%, (100-X=corresponding resin content)

In accordance with the present invention, epoxy resin composition consists of epoxy resin represented by formula (II), a naphthalene tetra-functional type epoxy resin, which has rigid naphthalene skeleton with high symmetry and high density aromatic rings which provides high melting and combustion temperature. This epoxy resin can provide the highest level of Tg in all kinds of epoxy resins and excellent curability. These features do assist in providing low flaming ability, low warpage and good solder cracking resistance.

When another epoxy resin is used in combination with the naphthalene tetra-functional type epoxy resin of formula (II), the other epoxy resin is an ordinary polymer having epoxy groups. There are cited, for example bisphenol type epoxy resins such as a resin of formula I, cresol novolac type epoxy resins, phenol novolac type epoxy resins, tri-functional epoxy resins (e.g., triphenolmethane type epoxy resins, and alkyl modified triphenolmethane type epoxy resins) and tri-azine nucleus-containing epoxy resins.

Paraxylene-modified phenolic resin curing agent (xyloc) with high density aromatic ring and bulky structure represented by formula (IV) tend to be thermally stable and possess higher flaming resistance as compared to a phenolic resin curing agent such as that of formula III. Thus, with above epoxy resin II and curing agent composition IV, lower filler loading to achieve UL-94 V-0 requirement is possible without additional external flame retardant agent. Filler loading should be 70% by weight or above based on total epoxy resin composition (with respective ratio of alumina to total filler calculated based on equation (II) and referring to example 2 and 3).

When the amount of filler is less than 70% by weight, the resulting alumina filler composition will be inadequate for flame retardant performance (comparative example 1), any of additional external flame retardant agents including bromine, antimony, boron, phosphorus or their compounds is needed to achieve UL-94 V-0. On the other hand, a filler content more than 76% but having a ratio of alumina to total filler content lower than respective amount calculated based on equation (II) also could not achieve UL-94 V-0 and additional external flame retardant agents are needed.

Comparative example 2 is filled with 80% filler content with a ratio of alumina filler to total filler content of 0.28, the resulting epoxy resin composition not able to achieve UL-94 V-0. However comparative example 3 which contain same composition as CE2 but added with 1.5% of antimony trioxide and 1.3% of brominated phenol novolac epoxy resin could achieved UL-94 V-0. When the amount of filler is more than 85% by weight, no alumina filler or additional external flame retardant agent is needed to achieve UL-94 V-0 (CE4).

The 2nd embodiment is represented by boundary equation (III) and illustrated in graph 1 where the UL-94 V-0 requirement can be achieved:

$$Y \geq -0.0686X + 5.77$$

equation (III)

Where,

Y = Ratio of alumina filler to total filler content (in wt%) is from 0 to 1.0

X = Total filler content in wt% is from 69.5% to 95%, (100-X=corresponding resin content)

In accordance with the present invention, the epoxy resin composition consists of the epoxy resin represented by formula (II) which is a naphthalene tetra-functional type epoxy resin, which has rigid naphthalene skeleton with high symmetry and high density aromatic rings provides high melting and combustion temperature. This epoxy resin can provide one of the highest level of Tg among various kinds of epoxy resins, excellent curability and relatively lower moisture absorption. These features are desirable in order to achieve good flaming ability, low warpage and good solder cracking resistance, all crucial requirements of new generation IC packaging materials.

Naphthalene resin curing agent with even higher density aromatic ring and the bulky structure is represented by formula (V). The weight fraction of hydrogen atoms is lower compared to paraxylene-modified phenolic resin curing agent (xyloc) represented by formula (IV). Hence it can possess higher flaming resistance as compared to xyloc resin curing agent. Thus, with above naphthalene type epoxy resin and curing agent composition, it allows lower filler loading epoxy resin composition to meet UL-94 V-0 requirement without additional flame retardant agent. Filler loading should be 69.5% by weight or above based on total epoxy resin composition (with respective ratio of alumina filler calculated based on equation (III) and referring to example 4 and 5.

When the amount of filler is less than 69.5% by weight, the resulting alumina filler composition will have inadequate flame retardant performance (CE5). On the other hand, a filler content more

than 69.5% but a ratio of alumina to filler content lower than the respective amount calculated based on equation (III) also could not achieve UL-94 V-0 and additional external flame retardant agents are needed. CE6 is filled with 80% filler content and ratio of alumina filler to total filler content is 0.2, the resulting epoxy resin composition is not able to achieve UL-94 V-0. However, CE7 which contain same composition as CE6 but added with 0.4% of antimony trioxide and 1.0% of brominated phenol novolac epoxy resin could achieved UL-94 V-0. When the amount of filler is more than 84% by weight, no alumina filler or additional external flame retardant agent is needed to achieve UL-94 V-0 (CE8).

When another epoxy resin is used in combination with the naphthalene tetra-functional type epoxy resin of formula (II), the other epoxy resin is an ordinary polymer having epoxy groups. There are cited, for example bisphenol type epoxy resins such as those of formula I, cresol novolac type epoxy resins, phenol novolac type epoxy resins, tri-functional epoxy resins (e.g., triphenolmethane type epoxy resins, and alkyl modified triphenolmethane type epoxy resins) and tri-azine nucleus-containing epoxy resins.

Graph 1 illustrates the present invention where the UL-94 V-0 requirement can be achieved with such composition of epoxy resin represented by equation, (II) and (III). It is within the scope of the invention for these boundary lines to be widened dependent on the self-flame retardant characteristics of the epoxy resin.

A typical filler distribution that can fulfill the above criterion should preferably be made up of three peaks in the regions of (i) 0.5 -3.5 μm , (ii) 5.0-20.0 μm and (iii) 20.0-100.0 μm with respective percentage of (i) 20-30%, (ii) 30-40% and (iii) 35-50% of the total filler composition. The lowest point in the 'saddle' region between 1.0-6.0 μm should be at least 0.5%, preferably more than 1.0% of the total epoxy resin composition.

The smallest peak (i) preferably comprises a fine silica filler that can be either fused or crystalline in nature. These fine fillers serve as lubricating 'bearings' that promote flowability as well as minimising air-vent flashes during transfer molding. Too much or too little of such filler can render these functions ineffective. In the present invention, peak (i) and the 'saddle' is delicately designed to account for 25-35% by wt of the total filler composition having maximum size less than 8 μm . Fillers exceeding 8 μm is not effective in performing the above mentioned functions. An alternative description to supplement the above can be given by $D_{10}=1.5\text{-}2.0\mu\text{m}$, $D_{50}=10\text{-}20\mu\text{m}$ and $D_{90}=70\text{-}80\mu\text{m}$, where DXX referring to diameter at XX% based on cumulative filler distribution.

The specific surface area of filler should be between 1.0-3.0 m^2/g . In addition, spherical fillers as well as wide filler distribution are desirable to maintain good flowability and processibility. In return, higher filler content loaded is possible. Spherical fillers also help in reducing abrasiveness and the modulus of the resin composition.

With regard to the blending ratio of the epoxy resin (A) to the phenolic resin curing agent (B), preferably the epoxy resin (A) is used in an amount of 0.8 to 1.25 epoxy equivalent per hydroxyl group equivalent of the phenolic resin curing agent (B).

The curing accelerator used in the present composition can be any material as long as it can accelerate the reaction between the epoxy group of the component (A) and the hydroxyl group of the component (B). There can be widely used those curing accelerators generally used in sealing materials, such as diazabicycloundecene (DBU), triphenylphosphine (TPP), benzyldimethylamine (BDMA), 2-methylimidazole (2-MZ) and the like. These compounds can be used alone or in combination of two or more.

The curing accelerator is preferably used in an amount of 0.1 to 1.0% by wt in the total epoxy resin composition. In an amount less than 0.1 wt %, hardness of the resulting molded product decreases and mold release characteristics tend to deteriorate. In an amount more than 1.0 wt %, the reaction proceeds undesirably fast and fluidity tends to decrease.

The present epoxy resin composition used for sealing comprises, as essential components, an epoxy resin, a curing agent, an inorganic filler and a curing accelerator. The resin composition may further comprise, if necessary, various additives such as silane coupling agent, releasing agent (e.g., natural wax or synthetic wax), stress-reducing additive (e.g., silicone oil or rubber), and the like.

In producing the present epoxy resin composition for sealing as a molding material, an epoxy resin, a curing agent, a curing accelerator, a filler and other additives are mixed sufficiently and uniformly using a mixer or the like; then, the mixture is melt kneaded using hot rolls, a kneader or the like; and the kneaded product is cooled and ground. The molding material thus obtained can be used for sealing, coating, insulation, etc. of electronic parts or electric parts.

According to the present invention, there can be obtained an environmental friendly resin composition having high thermal conductivity, low thermal expansion and low moisture absorption, among others. Such combination has not been attained with prior art.

DESCRIPTION OF PREFERRED EMBODIMENTS

The present invention is hereinafter described by way of the following Examples.

In the Examples, the amount of each component used is expressed as parts by weight. Example I uses a biphenyl epoxy resin of formula I which illustrates the molding processes and testing methods as used in Examples 2 and 3.

EXAMPLE 1

The following components in the following proportions are mixed at room temperature using a mixer a biaxial roll, cooled and ground to obtain a molding material. Epoxy compound of formula (I) (softening point: 108°C., epoxy equivalent: 185g/eq) 5.0 parts by weight
Phenol novolac resin curing agent, (softening point : 90°C., hydroxyl group equivalent: 105

g/eq)	2.9 parts by weight
Alumina filler	80.5 parts by weight
Fused silica filler	10.0 parts by weight
Diazabicycloundecene (DBU)	0.2 part by weight
Silane coupling agent	0.5 part by weight
Stress reducing additive	0.2 part by weight
Carbon black	0.3 part by weight
Releasing agent	0.4 part by weight

For epoxy resin composition flame retardant performance testing, UL-94, developed by Underwriters Laboratories, is one of the most widely used. It begins with pellet preparation, then mold into test piece with size at 125mm x 13mm x 3.2mm thickness. The molded test piece is then put into a chamber with temperature control at 23+/-2 °C and 50+/-5 °C relative humidity for at least 48hrs. After pre-treatment test piece is ready for burning test, which follow ASTM D5207. In

the test, the specimens are clamped vertically. A small $\frac{3}{4}$ in. high blue flame is applied to the bottom of the specimen for 10sec, withdrawn, and reapplied for an additional 10 sec; the duration of flaming and glowing is noted as soon as the specimen has extinguished. A layer of cotton is placed beneath the specimen to determine whether dripping materials will ignite it during the test period. Five test pieces are needed for every sample. If total flaming time of five test pieces is less than 50 seconds, the epoxy resin composition is considered to pass UL-94 test and classified as V-0. On the other hand, if total flaming time is more than 50 seconds, the sample is considered fail UL-94 test and classified as V-1 and V-2.

The materials are classified V-2, the least stringent classification, or V-1 or V-0, which are the most stringent classification for the test. Table 1 below summarizes the requirements for each classification.

Performance testing at the package level begins with pellet preparation into various sizes. Depending on test purposes, it can be molded into 35x35mm Over-Molded-Plastic Ball-Grid-Array (OMPGBGA) with 352 balls on an organic substrate with solder resist at a temperature of 175°C, 50-80kgf/cm² and 60sec. Subsequently the packages with a chip in it (9x9mm, polyimide(PI) coated) are exposed to an atmosphere of 60°C/60% RH for 120hrs. Thereafter the packages are sent through a varying temperature profile according to the JEDEC standard with a peak temperature of 255°C in an infrared (IR) oven. Thereafter, cracks present at the surfaces of the devices are observed using a microscope. In addition, the packages are also scanned using CSAM (C-mode Scanning Acoustic Microscope) for delaminated interfaces with focus on the chip top and the substrate-mold compound (iii) with these chips to obtain devices for solder cracking test. The tablets are also made into 16p DIPs (16-pin Dual-Inline Packages) for HTSL test at 175°C, after which circuit resistance are examined for values exceeding 1.0Ω (value of electrical resistance after mold is typically <0.80Ω. During prolonged exposure to high temperature, free ions present within the encapsulant material, especially bromide and antimony can corrode the gold ball bond to form for example Kirkendall void which in turn leads to higher electrical resistance). The results are expressed by average life.

For thermal conductivity measurement the epoxy resin composition is first made into tablets and then into the cylindrical blocks with diameter 40mm by height 25±5mm at the temperature of 175°C, 150kgf/cm² and 300sec using a low pressure transfer molding machine.

Table 1 :Summary of UL 94 Vertical Burning Test for Classification Materials V-0, V-1, V-2

Criteria	Classification		
	V-2	V-1	V-0
Number of specimens	5	5	5
Number of Ignitions	2	2	2
Maximum flaming time per specimen per flame application (sec)	30	30	10
Total flaming time, five specimens, 2 ignitions (sec)	250	250	50
Flaming drips ignite cotton	Yes	No	No
Maximum afterflow time, per specimen (sec)	60	60	30
Burn to holding clamp	No	No	No

The following results were achieved:

Thermal conductivity (W/mK)	3.0
UL-94 / 3.2mm (V-0/V-1)	V-0
Soldering crack test (PBGA-35x35/PI)	0/4

(no. of cracked devices/total no.)

Delamination check (PBGA-35x35/PI)

60°C/60% RH for 120hrs

0/4

HTSL at 175°C (hr to 1st failure)

1000

Examples 2-3

Epoxy resin compositions are compounded according to recipes of 1st embodiment shown in Table 2, and subjected to the same procedures as in Example 1 to obtain molding materials. Test results for these epoxy resin compositions were obtained in the same manner as in Example 1. The results are shown in Table 2.

Examples 4-5

Epoxy resin compositions are compounded according to recipes 2nd embodiment shown in Table 3, and subjected to the same procedures as in Example 1 to obtain molding materials. Test results for these epoxy resin compositions were obtained in the same manner as in Example 1. The results are shown in Table 3.

Comparative Examples 1-8

Additional epoxy resin compositions are compounded based on recipes outside the claimed region and labelled comparative examples in Table 2, 3 and 4. These epoxy resin compositions are subjected to the same procedures as in Example 1 to obtain molding materials. Test results for these molding materials were obtained in the same manner as in Example 1. The results are shown in Table 2 and 3.

Table 2 (1st Embodiment)

	Examples		Comparative Examples			
	2	3	CE1	CE2	CE3	CE4
Alumina filler	30.0	38.0	65.0	22.0	22.0	0.0
Fused silica filler	50.0	40.0	3.0	58.0	58.0	89.0
Alumina filler:Filler content (wt %)*	0.38	0.49	0.96	0.28	0.28	0.00
Naphthalene epoxy resin	11.3	12.0	17.0	11.3	9.5	6.0
Paraxylene-modified phenolic curing agent	7.0	8.3	11.0	7.0	6.0	3.3
Diazabicycloundecene (DBU)	0.3	0.3	1.5	0.3	0.3	0.3
Silane coupling agent	0.5	0.5	1.0	0.5	0.5	0.5
Stress reducing additive	0.2	0.2	0.2	0.2	0.2	0.2
Carbon black	0.3	0.3	0.3	0.3	0.3	0.3
Releasing agent	0.4	0.4	1.0	0.4	0.4	0.4
Antimony trioxide	0.0	0.0	0.0	0.0	1.5	0.0
Brominated phenol novolac epoxy resin	0.0	0.0	0.0	0.0	1.3	0.0
Thermal conductivity (W/mK)	1.3	1.1	1.1	1.0	1.0	1.4
UL-94 / 3.2mm (V-1/V-1)	V-0	V-0	V-1	V-1	V-0	V-0
Soldering crack test (PBGA-35x35/PI)	0/4	0/4	0/4	0/4	0/4	0/4
(no. of cracked devices/total no.)						
Delamination check (PBGA-35x35/PI)						
60°C/60% RH for 120hrs	0/4	1/4	1/4	0/4	1/4	0/4
HTSL at 175°C (hr to 1 st failure)	800	800	900	500	500	1000

* : the ratio of Alumina filler (in wt %) to total filler content (FC in wt%)

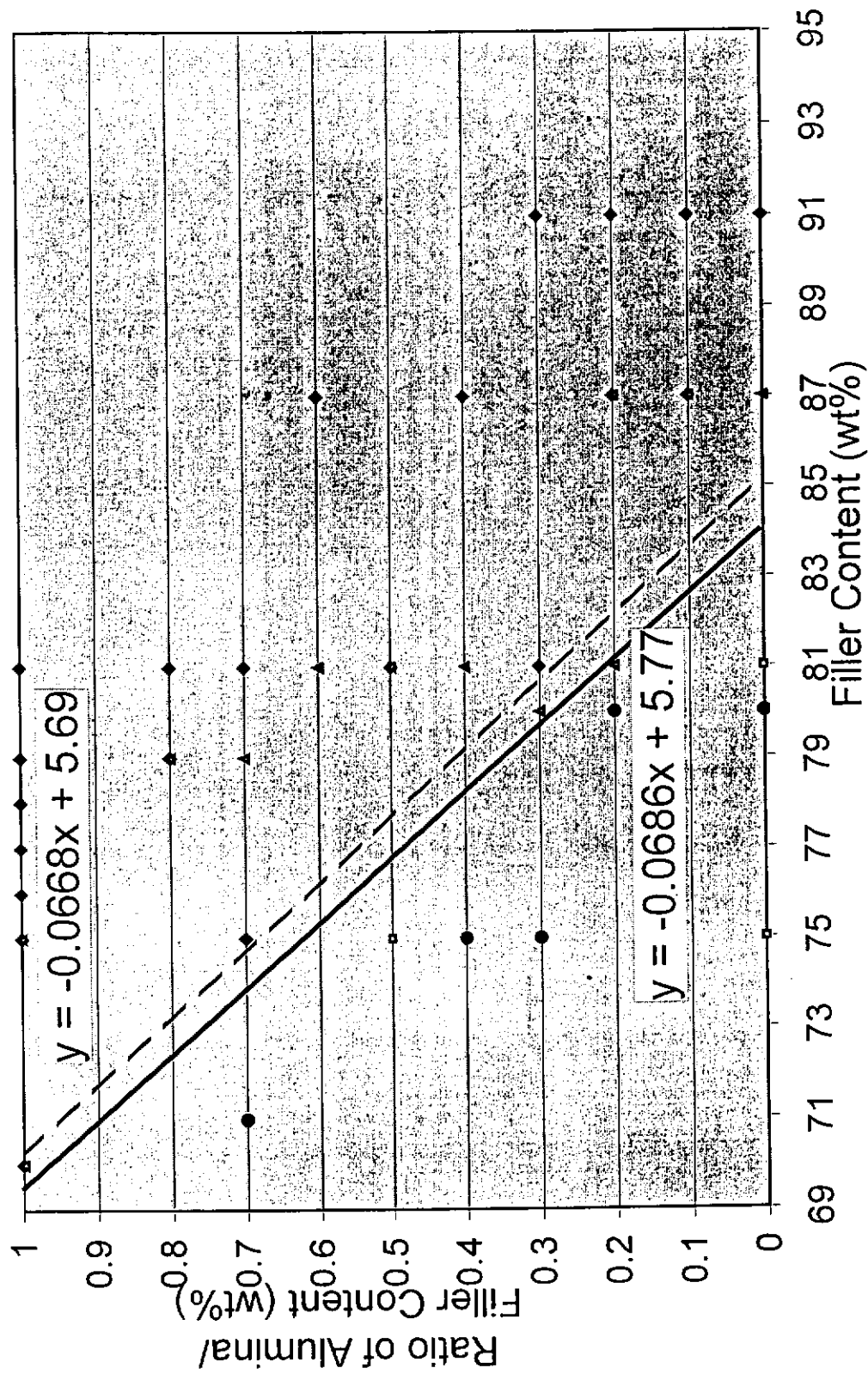
Table 3 (2nd Embodiment)

	Examples		Comparative Examples			
	4	5	CE5	CE6	CE7	CE8
Alumina filler	39.0	53.0	60.0	16.0	16.0	0.0
Fused silica filler	39.0	22.0	8.0	64.0	64.0	87.0
Alumina filler:Filler content (wt %)*	0.50	0.71	0.88	0.20	0.20	0.00
Naphthalene epoxy resin	9.7	11.0	13.0	8.8	8.1	5.2
Naphthalene curing agent	10.5	12.2	15.0	9.5	8.8	6.2
Diazabicycloundecene (DBU)	0.4	0.4	1.5	0.3	0.3	0.2
Silane coupling agent	0.5	0.5	1.0	0.5	0.5	0.5
Stress reducing additive	0.2	0.2	0.2	0.2	0.2	0.2
Carbon black	0.3	0.3	0.3	0.3	0.3	0.3
Releasing agent	0.4	0.4	1.0	0.4	0.4	0.4
Antimony trioxide	0.0	0.0	0.0	0.0	0.4	0.0
Brominated phenol novolac epoxy resin	0.0	0.0	0.0	0.0	1.0	0.0
Thermal conductivity (W/mK)	1.5	1.5	0.9	1.0	1.0	1.1
UL-94 / 3.2mm (V-1/V-1)	V-0	V-0	V-1	V-1	V-0	V-0
Soldering crack test (PBGA-35x35/PI) (no. of cracked devices/total no.)	0/4	0/4	0/4	0/4	0/4	0/4
Delamination check (PBGA-35x35/PI) 60°C/60% RH for 120hrs	1/4	0/4	1/4	0/4	2/4	0/4
HTSL at 175°C (hr to 1 st failure)	800	900	900	500	500	1000

* : the ratio of Alumina filler (in wt %) to total filler content (FC in wt%)

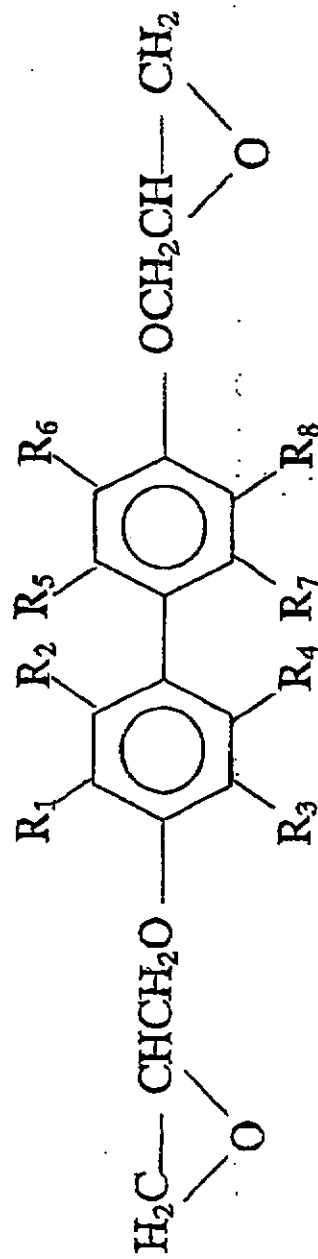
As used herein the term "comprising" is used in an inclusive sense such that other components can be present in the composition as well as those specifically referred to.

Boundary line



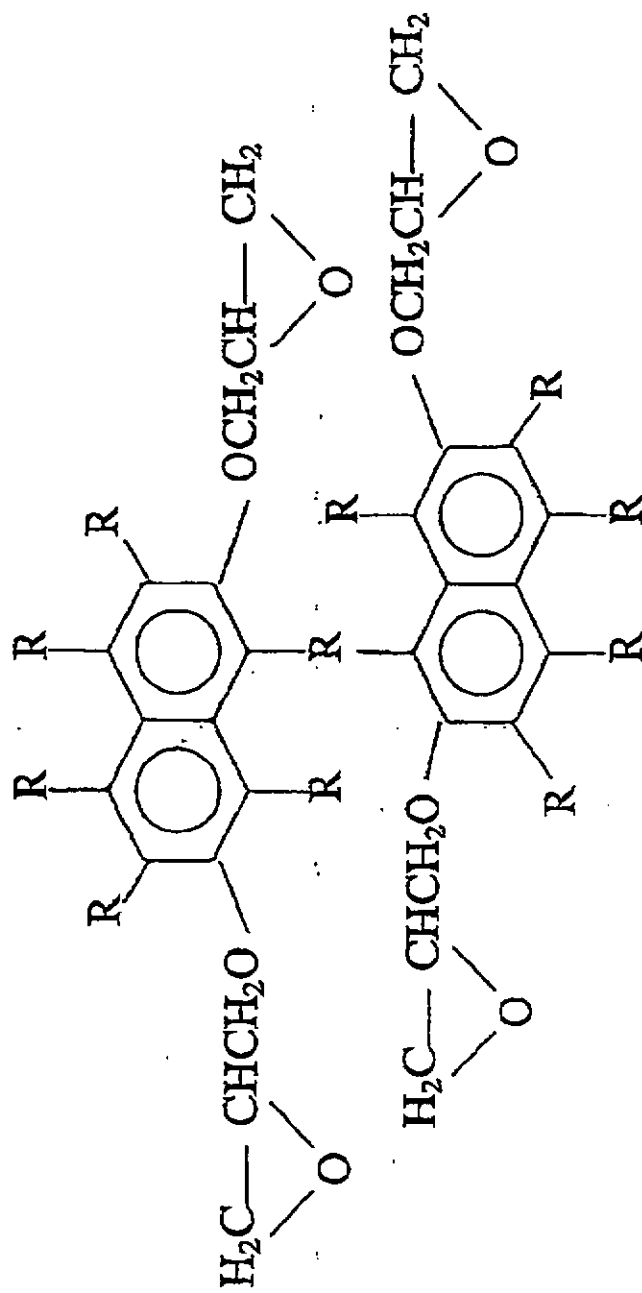
Graph 1: Boundary line represented by equation (II) and (III)

Formula (I)



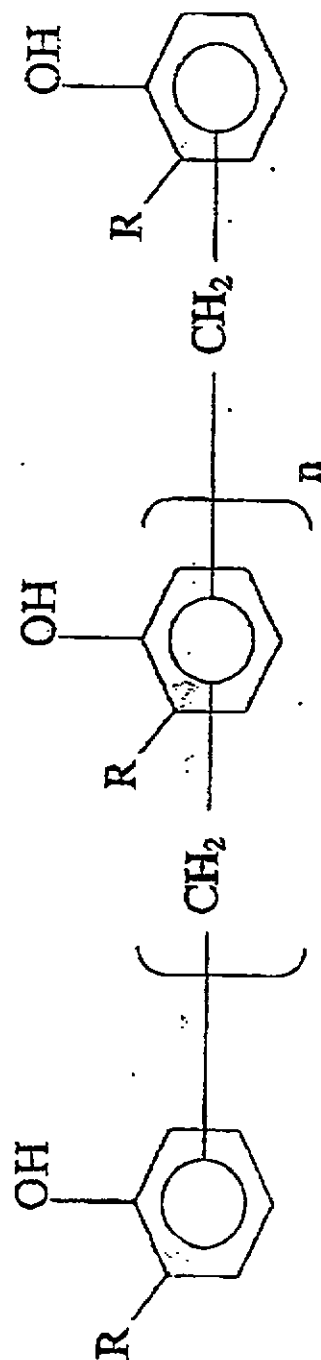
Wherein R_1 to R_8 is independently an atom or group selected from hydrogen atom, a halogen and an alkyl group

Formula (II)



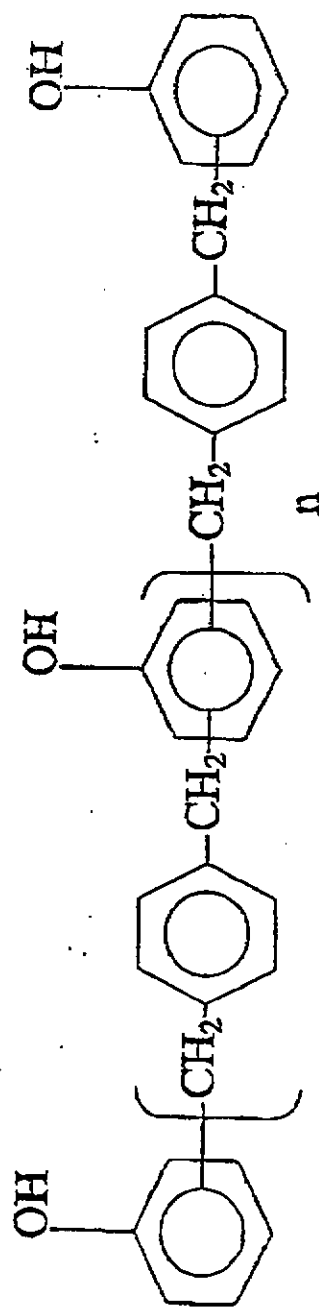
Wherein R is independently an atom or group selected from hydrogen atom, a halogen and an alkyl group

Formula (III)



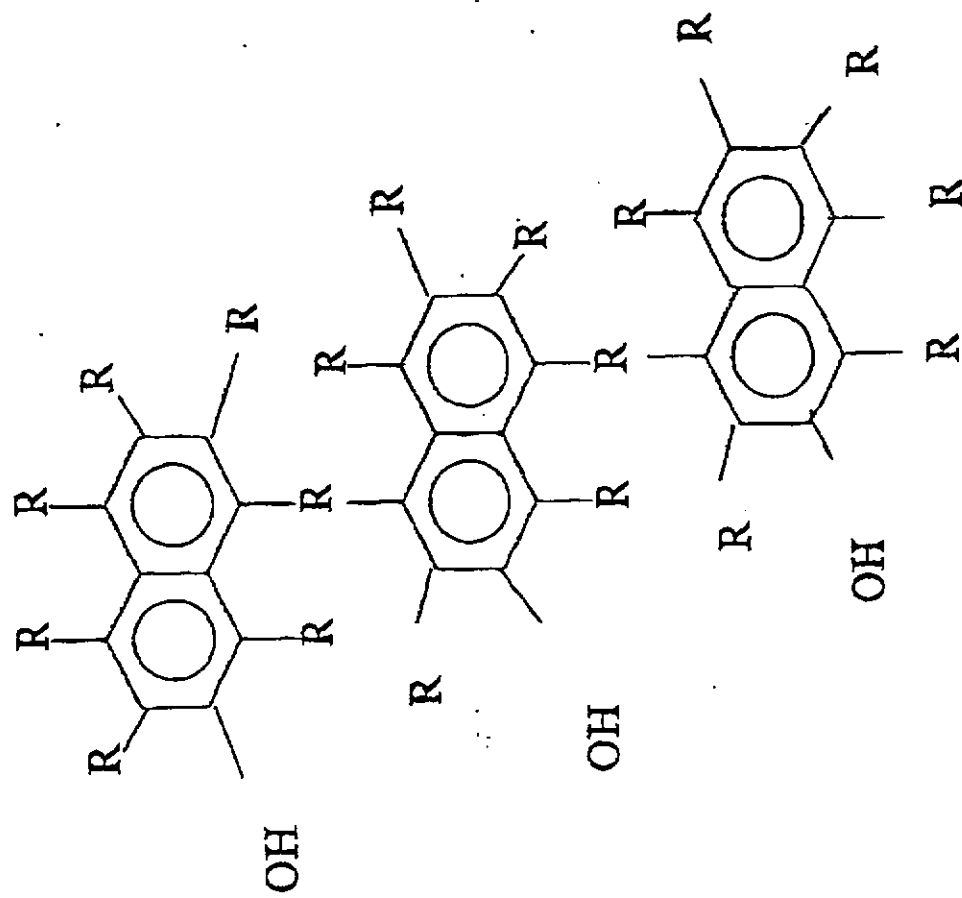
Wherein n is an integer of 0-10 and each R is independently an atom or group selected from hydrogen atom, a halogen and an alkyl group

Formula (IV)



Wherein n is an integer of 0-5 and $n=0: 0.07$, $n=1: 0.25$, $n=2: 0.38$, $n=3: 0.14$, $n=4: 0.12$ and $n=5: 0.04$

Formula (V)



Wherein R is independently an atom or group selected from hydrogen atom, a halogen and an alkyl group

CLAIMS:

1. An epoxy resin composition used for sealing of semiconductor devices, comprising:
 - (A) an epoxy resin of the formula II,
 - (B) an epoxy resin curing agent,
 - (C) a mixture of inorganic aluminum oxide and silicon oxide constituting 69.5-95% by weight of the total epoxy resin composition,
 - (D) a curing accelerator
2. An epoxy resin composition of claim 1 wherein the epoxy resin (A) is used in an amount of 0.8 to 1.2 times the epoxy equivalents per hydroxyl group equivalent of said phenolic resin (B).
3. An epoxy resin composition of claim 1 or claim 2 wherein the curing accelerator (D) is present in an amount from 0.1-1 weight per cent based on the total resin composition.
4. An epoxy resin composition according to claim 1 used for encapsulating semiconductor devices.
5. The epoxy resin composition of claim 1, wherein the amount of the mixture of aluminum oxide and silicon oxide is defined by equation II:

$$Y \geq -0.0668X + 5.69$$

Where,

Y = Ratio of alumina filler to total filler content (in wt%) is from 0 to 1.0

X = Total filler content in wt% is from 69.5% to 95%, (100-X=corresponding resin content)

6. The epoxy resin composition of claim 5, wherein the naphthalene tetra-functional type resin containing 5-30% by weight represented by formula (II) is present in an amount of from 5 to 50% by weight, based on total epoxy resin composition.
7. The epoxy resin composition of claim 5 or 6, wherein the paraxylene-modified phenolic resin curing agent (xyloc) represented by formula (IV), is present in an amount of from 50 – 100% by weight based on total resin curing agent composition in epoxy resin composition.
8. The epoxy resin composition of claim 1, the amount of the mixture of aluminum oxide and silicon oxide is defined by equation III:

$$Y \geq -0.0686X + 5.77$$

Where,

Y = Ratio of alumina filler to total filler content (in wt%) is from 0 to 1.0

X = Total filler content in wt% is from 69.5% to 95%, (100-X=corresponding resin content)

9. The epoxy resin composition of claim 8, where in the naphthalene tetra-functional type resin represented by formula (II) is present in an amount of from 5 to 30.5% by weight based on total epoxy resin composition.

10. The epoxy resin composition of claim 11, wherein the naphthalene resin curing agent represented by formula (V) is present in an amount of from 80 to 100% by weight based on total resin curing agent composition in epoxy resin composition.