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(54) Title: EPOXY MOLDING COMPOUND, PREPARATION AND USE THEREOF

(57) Abstract: An epoxy molding compound, preparation and use thereof are provided. The epoxy molding compound comprises an epoxy resin, a phenolic resin, a curing accelerator, a specific crystalline silica optionally in combination with a specific spherical silica, and one or more additives selected from the group consisting of a flame retardant, a wax, a coupling agent, a pigment, an ion trapping agent and a stress absorbing agent. The epoxy molding compound has a high thermal conductivity and a low wire sweep while maintaining other desired properties as required in the full-molded modules.



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Epoxy Molding Compound, preparation and use thereof

Technical field

5 The present invention relates to an epoxy molding compound comprising a crystalline silica having a specific particle size distribution optionally in combination with a spherical silica having a specific particle diameter, said epoxy molding compound has a high thermal conductivity and a low wire sweep. The present invention also relates to a preparation method of the epoxy molding compound and
10 use of the epoxy molding compound for full-molded modules.

Background art

Epoxy molding compound ("EMC") is widely used in various encapsulating fields such as semiconductor devices, integrated circuit, consuming electronics, vehicles and the like. EMC plays a very important role in the electronics encapsulation, such
15 as protecting the devices inside against influences such dust, moisture, radiation or mechanical impact and assisting heat dissipation.

With the development of microelectronics packaging technology, EMC, as a dominating microelectronics packaging material, develops fast as well, and possesses more than 90% market share of the microelectronics packaging
20 materials.

Molded epoxy resin products are produced from epoxy molding compounds. A typical epoxy molding compound comprises one or more epoxy resins, one or more curing agents (hardener), one or more curing accelerators (catalyst), one or more fillers and optionally one or more additives. An epoxy molding compound can be
25 molded and cured to a solid shaped article in a mold at an elevated temperature and for a certain time. Afterwards, the demolded article is usually post-cured at an elevated temperature to complete the curing reaction and obtain a resin article with the desired properties.

US8362115B2 discloses an epoxy resin composition comprising a specific epoxy resin, a phenolic resin, a cure accelerator and an inorganic filler. The epoxy resin
30 composition disclosed herein has a better reliability and better warpage properties.

But US8362115B2 is silent about the properties of thermal conductivity and wire sweep.

Modules are advanced hybrid integrated semiconductors, such as integrated circuit and power devices. Traditional modules are semi-encapsulated, which enables high power density and fast heat dissipation and requires simple encapsulation processes. However, such a semi-encapsulation has disadvantages of low anti-jamming ability, low safe coefficient, low resistance to high voltage and high humidity. Full-molded modules mean that all semiconductors are encapsulated within the EMC without bare radiators, which improves electrical performances and stability of the modules. The full-molded modules are a development tendency of the modules.

In the full-molded modules, it has higher requirements for the thermal conductivity and flowability for the EMC. High thermal conductivity always means high loading of high thermal conductivity filler(s), so traditional high thermal conductivity EMC for full-molded TO application always requests high viscosity, which, however, may lead to a reduction of flowability and an increase of the wire sweep of the full-molded modules since many functional units are integrated within one module.

Therefore, it still needs an epoxy molding compound that may balance the high thermal conductivity and high flowability and still maintain other desired properties as required in the full-molded modules.

Summary of the invention

In one aspect, the present invention provides an epoxy molding compound having both a high thermal conductivity and a low wire sweep while maintaining other properties as required in the full-molded modules.

The epoxy molding compound of the present invention comprises:

- (a) an epoxy resin,
- (b) a phenolic resin,
- (c) a curing accelerator,
- (d) crystalline silica, optionally in combination with spherical silica, and
- (e) one or more additives selected from the group consisting of a flame retardant, a wax, a coupling agent, a pigment, an ion trapping agent and a stress absorbing agent;

wherein the particle size distribution of the crystalline silica is that 5-50 wt.%, preferably 5-45 wt.% of the crystalline silica is less than 10 μm and 50-95 wt.%, preferably 55-95 wt.% of the crystalline silica is greater than 10 μm but less than 75 μm ,

5 the spherical silica has a particle diameter in the range of from 0.1 to 30 μm , preferably from 0.1 to 20 μm , more preferably 0.1 to 10 μm .

the total amount of the crystalline silica and the spherical silica (if present) is about 40-90 wt.%, preferably about 50-88 wt.%, based on the total weight of the composition,

10 the weight ratio between the crystalline silica and the spherical silica (if present) is in a range of from 5:1 to 35:1, preferably from 10:1 to 30:1, more preferably from 12:1 to 28:1, and

the molar ratio between the hydroxyl groups in the phenolic resin and the epoxy groups in the epoxy resin is in a range of from 0.5 to 2, preferably from 0.8 to 1.7, more preferably from 0.9 to 1.5.

Another aspect the present invention is to provide a method for preparing the epoxy molding compound of the present invention, comprising steps of:

(1) weighing up all of the components and mixing them in a high speed mixer to get a premixed powder; and

20 (2) extruding the premixed powder by an extruder to get an extrusion product and then crushing the extrusion product into powder.

The extruder can be a twin-screw extruder. The extrusion may preferably be performed at an elevated temperature, such as a temperature in the range of from 60°C to 110°C.

25 Yet another aspect of the present invention is the use of the epoxy molding compound of the present invention for full-molded modules.

Detailed description

The present invention will be described in detail as follows. The materials, methods, and examples herein are illustrative only and, except for specifically stated otherwise, are not intended to be limiting. Suitable methods and materials are described herein, 30 although methods and materials similar or equivalent to those described herein can

be used in the practice or testing of the present invention. All publications and other references mentioned herein are explicitly incorporated by reference in their entirety.

Unless otherwise defined, all technical and scientific terms used herein have the same meanings as commonly understood by those skilled in the art. In case of
5 conflict, the present specification, including definitions, is decisive.

Unless stated otherwise, all percentages, parts, ratios, etc., are by weight.

Where a range of numerical values are recited herein, unless otherwise stated, the range is intended to include the endpoints thereof, and all integers and fractions within the range.

10 Use of “a” or “an” is employed to describe elements and components of the present invention. This is done merely for convenience and to give a general sense of the invention. This description should be read to include one or at least one and the singular also includes the plural unless it is obvious that it is meant otherwise.

15 The terms “comprising”, “comprises” and “comprised of” as used herein are synonymous with “including”, “includes” or “containing”, “contains”, and are inclusive or open-ended and do not exclude additional, non-recited members, elements or method steps.

Other than in the operating examples, or where otherwise indicated, all numbers expressing quantities of ingredients, reaction conditions, or defining ingredient
20 parameters used herein are to be understood as modified in all instances by the term “about”.

The term “room temperature (RT)” used herein refers to around 25°C.

Each component in the epoxy molding compound of the present invention will be described in detail below.

25 Epoxy resin (a)

As used herein, the term “epoxy resin” means a polymer containing generally two or more epoxide groups per molecule.

In the epoxy molding compound of the present invention, there are no particular

limitations for the epoxy resins and any epoxy resins commonly used are suitable. Examples of the epoxy resins usable in the present invention include: bisphenol resins such as bisphenol A epoxy resins and bisphenol F epoxy resins; biphenyl epoxy resins such as biphenyl epoxy resins and tetramethylbiphenyl epoxy resins; 5 novolac epoxy resins such as phenol novolac epoxy resins, cresol novolac epoxy resins, bisphenol A novolac epoxy resins, epoxy compounds of condensates of phenols and phenolic hydroxyl group-containing aromatic aldehyde, and biphenylnovolac epoxy resins; triphenylmethane epoxy resins; tetraphenylethane epoxy resins; dicyclopentadienephenol epoxy resins; phenolaralkyl epoxy resins; 10 epoxy resins each having a naphthalene skeleton in its molecular structure, such as naphthol novolac epoxy resins, naphtholaralkyl epoxy resins, naphthol-phenol co-condensated novolac epoxy resins, naphthol-cresol co-condensated novolac epoxy resins, diglycidylloxynaphthalene; and 1,1-bis(2,7-diglycidylloxy-1-naphthyl)-alkanes. Any of these may be used alone or in combination of two or more.

15 Preferably, the epoxy resin used in the present invention can be one or more selected from o-cresol novolac epoxy resins (EOCN), dicyclopentadienephenol epoxy resins (DCPD), biphenyl epoxy resins (BP), bisphenol A novolac epoxy resins (Bis-AN), triphenylmethane epoxy resins, epoxy resins having a naphthalene skeleton, multi-aromatic epoxy resins (MAR) and multi-functional epoxy resins 20 (MFN).

The amount of the epoxy resin in the composition of the present invention may be preferably 1 to 25 wt.%, more preferably 5 to 20 wt.%, most preferably 5 to 15 wt.%, based on the total weight of the composition.

Phenolic resin (b)

25 The phenolic resin of the present invention is mainly used as a curing agent (hardener). The hydroxyl groups contained in the phenolic group react with the epoxy groups of the epoxy resin to form a network structure.

Phenolic resins can be used in the composition of the present invention without particular limitation as long as the phenolic resins are conventionally used in the 30 EMC and do not do harm to the desired effect of the present composition. Examples of the phenolic resin that can be used in the present invention include phenol novolac

resins, cresol novolac resins, aromatic hydrocarbon formaldehyde resin-modified phenol resins, dicyclopentadienephenol addition-type resins, phenol aralkyl resins (Xylok resin), naphthol aralkyl resins, trimethylolmethane resins, tetraphenylethane resins, naphthol novolac resins, naphthol-phenol co-condensated novolac resins, naphthol-cresol co-condensated novolac resins, biphenyl-modified phenol resins (having phenol nuclei connected through a bismethylene group), biphenyl-modified naphthol resins (having naphthol nuclei connected through a bismethylene group), aminotriazine-modified phenol resins (having phenol nuclei connected through melamine or benzoguanamine), alkoxy group-containing aromatic ring-modified novolac resins (having phenol nuclei and alkoxy group-containing aromatic rings connected through formaldehyde). The above phenolic resins can be used alone or in a combination of two or more in the composition of the present application.

Among these, the phenolic resin used in the composition of the present application can preferably be one or more selected from phenol novolac resins, Xylok resins, multi-aromatic phenolic resins and multi-functional phenolic resins.

The amount of the phenolic resin in the composition of the present invention may be preferably 1 to 20 wt.%, more preferably 2 to 15 wt.%, most preferably 2 to 10 wt.%, based on the total weight of the composition.

Curing accelerator (c)

As used herein, the term "curing accelerator" has the same meaning as "catalyst", which catalyzes or accelerates the curing reaction between the epoxy resin and the hardener.

Various compounds, such as organophosphorus compounds, amines, amidine compounds, imidazole compounds, organic acid metal salts, Lewis acids, amine complex salts and so on can be included in the epoxy molding compound as a curing accelerator.

Preferably, the curing accelerator used in the composition of the present application is one or more selected from organophosphorus compounds, imidazole compounds, amines, and amidine compounds. More preferably, specific examples of the curing accelerator used in the composition of the present application include triphenyl phosphine (TPP), 1,8-diazabicyclo(5,4,0) undecene-7 (DBU), 2,4-diamino-6[2'-methylimidazolyl-(1')]ethyl-s-triazine (2MZ), 2-phenyl-4-methyl imidazole (2P4MZ),

N,N-dimethyl benzyl amine and triethylamine. The above specific curing accelerators may be used alone or in a combination of two or more in the composition of the present application.

The more the amount of the curing accelerator is, the faster the curing speed is. But excess curing accelerator may cause short curing time and short spiral flow of the EMC. The amount of the curing accelerator in the composition of the present invention may be preferably 0.01 to 5 wt.%, more preferably 0.01 to 2 wt.%, most preferably 0.05 to 1 wt.%, based on the total weight of the composition.

Crystalline silica and spherical silica (d)

Silica, as a filler in the EMC, constitutes 60 wt% to 90 wt % of the total weight of the composition, and hence, the selection of the silica will greatly influence the properties of the EMC.

In the EMC, main roles of the silica are to improve thermal conductivity, reduce coefficient of thermal expansion and shrinkage of the EMC, and improve reliability. In order to achieve the above advantages, the filling rate of silica should be as high as possible. However, too much high filling rate of silica may increase viscosity, reduce the flowability of the composition and reliability of the molded products, and increase the wire sweep.

After intensive studying, the inventors of the present invention found that by selecting a specific type of silica, a specific combination of silicas and specific particle size distribution of the silica, compared with compositions of the prior art, the thermal conductivity, flowability and reliability can be improved and the wire sweep can be reduced.

In an embodiment of the present invention, the epoxy molding compound comprises a crystalline silica having a particle size distribution that 5-50 wt.%, preferably 5-45 wt.% is less than 10 μm and 50-95 wt.%, preferably 55-95 wt.% is greater than 10 μm but less than 75 μm .

In a preferred embodiment of the present invention, the epoxy molding compound comprises a combination of a crystalline silica and a spherical silica, wherein the crystalline silica having a particle size distribution that 5-50 wt.%, preferably 5-45

wt.% is less than 10 μm and 50-95 wt.%, preferably 55-95 wt.% is greater than 10 μm but less than 75 μm , the spherical silica has a particle diameter in the range of from 0.1 to 30 μm , preferably from 0.1 to 20 μm , more preferably from 0.1 to 10 μm , and the weight ratio between the crystalline silica and the spherical silica is in a range of from 5:1 to 35:1, preferably from 10:1 to 30:1, more preferably from 12:1 to 28:1.

The total amount of the silica(s) in the composition of the present invention may be 40 to 90 wt.%, preferably 50 to 88 wt.%, more preferably 60 to 85 wt.%, based on the total weight of the composition.

Other additives (e)

The epoxy molding compound of the present invention may further comprise one or more other additives selected from the group consisting of flame retardants, waxes, coupling agents, pigments, ion trapping agents, and stress absorbing agents. The additive(s) used in the composition of the present invention is/are not particularly limited as long as it/they does/do no harm to the desired effect(s) of the composition of the present invention.

As for the flame retardant, any suitable flame retardants can be used. However, from the view point of environment issue, a non-halogen flame retardant is preferable; an inorganic flame retardant is more preferable. Examples of the inorganic flame retardant include metal hydroxides, metal oxides, boron compounds, calcium silicate, and the like. Examples of the metal hydroxides include aluminum hydroxide, magnesium hydroxide, dolomite, hydrotalcite, calcium hydroxide, barium hydroxide, zirconium hydroxide, and the like. Examples of the metal oxides include zinc molybdate, molybdenum trioxide, molybdenum oxide, zinc stannate, tin oxide, aluminum oxide, iron oxide, titanium oxide, manganese oxide, zirconium oxide, zinc oxide, cobalt oxide, bismuth oxide, chromium oxide, nickel oxide, tungsten oxide, antimony trioxide, and the like. Examples of the boron compounds include zinc borate, zinc metaborate, barium metaborate, boric acid, borax and the like. The inorganic flame retardant can be used in the composition of the present application alone or in a combination of two or more. If required, organic flame retardant(s) such as brominated epoxy resins can be used in the composition of the present invention alone or in combination of the inorganic flame retardant(s). The amount of the flame

retardant in the composition of the present invention may be preferably 0 to 20 wt.%, more preferably 3 to 15 wt.%, most preferably 3 to 10 wt.%, based on the total weight of the composition.

The wax is used as a releasing agent in the composition of the present invention, and all suitable waxes can be used as long as the desired effect of the composition of the present invention is not affected. Examples of the wax include natural waxes and synthetic waxes. Preferred examples include montan waxes, fatty acid ester waxes, fatty acid waxes, aliphatic ester waxes, polyethylene waxes, polypropylene waxes, alkyl oligomer waxes and amide waxes. The wax can be used in the composition of the present application alone or in a combination of two or more. The amount of the wax in the composition of the present invention may be preferably 0 to 5 wt.%, more preferably 0.2 to 5 wt.%, most preferably 0.3 to 3 wt.%, based on the total weight of the composition.

The coupling agent of present invention, if present, improves the compatibility between the epoxy resin and the silica and increases the adhesion of the epoxy resin to a specific surface, such as a chip. The coupling agent is preferably selected from compounds comprising at least one oxirane group and at least one group, which is capable of interacting with the inorganic filler material. Examples of the coupling agent used in the composition of the present invention include silane coupling agents such as epoxysilanes, aminosilanes, mercaptosilanes, ureidosilanes, vinylsilanes, alkylsilanes. The coupling agent can be used in the composition of the present application alone or in a combination of two or more. The amount of the coupling agent in the composition of the present invention may be preferably 0 to 5 wt.%, more preferably 0.2 to 5 wt.%, most preferably 0.2 to 3 wt.%, based on the total weight of the composition.

Pigment in the composition of the present invention may be used to distinguish/mark different types of the devices and cover the design of the encapsulated units and prevent light from passing through. The epoxy molding compound of the present invention may comprise one or more pigments according to the practical needs. Example of the pigment includes carbon black. The amount of the pigment in the composition of the present invention may be preferably 0 to 5 wt.%, more preferably 0.2 to 5 wt.%, most preferably 0.4 to 3 wt.%, based on the total weight of the

composition.

The ion trapping agent is used in the composition of the present invention to reduce the content of free ions in the epoxy molding compound. The ion trapping agents commonly used in the art may be used in the composition of the present invention without particular limitation. Examples of the ion trapping agent used in the composition of the present invention include magnesium aluminum hydroxide type, aluminosilicate, antimony hydroxide type and bismuth hydroxide type. The ion trapping agent can be used in the composition of the present application alone or in a combination of two or more. The amount of the ion trapping agent in the composition of the present invention may be preferably 0 to 5 wt.%, more preferably 0.1 to 3 wt.%, most preferably 0.1 to 1 wt.%, based on the total weight of the composition

The stress absorbing agent is used in the composition of the present invention to reduce the elastic modulus and increase flexibility and reduce the coefficient of thermal expansion. Examples of the stress absorbing agent include silicone oil, silicone resins, butadiene-type rubbers and the like. The amount of the stress absorbing agent in the composition of the present invention may be preferably 0 to 5 wt.%, more preferably 0.1 to 2 wt.%, most preferably 0.1 to 1 wt.%, based on the total weight of the composition.

Depending on desired properties, the relative proportions of the individual components may vary within comparatively wide limits. The additives can be formulated into the epoxy molding compound as desirable.

There is no special limit for the preparation method of the epoxy molding compound of the present invention. In a preferred embodiment, the epoxy molding compound is manufactured by a process comprising steps of:

(1) weighing up all of the components (a) to (e) and mixing them in a high speed mixer at 50~500rpm, preferably 100~400rpm to get a premixed powder; and

(2) extruding the premixed powder by a twin-screw extruder to get an extrusion product at 60~110°C with the rotation speed of the paddle being 50~250 rpm and then crushing the extrusion product into powder.

Examples

The present invention will be further described and illustrated in details with

reference to the following examples, which, however, are not intended to restrict the scope of the present invention. All numbers in the examples are expressed in parts by weight.

Examples 1-10 and comparative examples 1-2

5 The raw materials used for the epoxy molding compound of each of Examples 1-10 according to the present invention and Comparative Examples 1 and 2 were weighed up as shown in Table 1. All the raw materials were mixed by a high speed mixer for 15 minutes at 250r/min below 10 °C to get a premixed powder. The premixed powder was then extruded by a twin-screw extruder at about 100 °C with the rotation speed
10 of the paddle being 200rpm. The extruded material was obtained and crushed into powder.

Various properties of the obtained epoxy molding compounds were tested according to the following test methods, and the results are shown in Table 1 as follows.

Test methods

Spiral flow:

In the spiral flow test the flow property of the epoxy molding compound was determined by measuring the length and weight of the resin flowing along the path of a spiral cavity. Sample for the spiral flow test was the powder sample of the epoxy molding compound. No additional preparation was required. The spiral flow test was
20 done according to the method EMI-1-66. Test conditions were set as follows: temperature 175 °C, pressure 70km/cm² and cure time 90s.

An acceptable spiral flow length of the EMC is in the range of from 15 inch to 65 inch, preferably from 20 inch to 55 inch.

Gel Time:

25 In the gel time test the gelation point of the epoxy molding compound was tested. In the test, a hot plate was heated to the temperature of 175°C. The powder sample of the epoxy molding compound was placed on the hot plate and let it stand as long as the sample was gelled, with stopwatch gelling time was measured (stopwatch was started immediately when the sample was placed on the hot plate and stopped when

gelling was complete).

An acceptable gel time of the EMC is in a range of from 15 sec to 60 sec, preferably from 20 sec to 50 sec.

Viscosity:

- 5 The Viscosity value was determined using a capillary rheometer CFT-500D from SHIMADU, the test temperature was 175 °C and the load was 10 Kgf.

Thermal conductivity:

- 10 The thermal conductivity value of the molded composition was determined using a thermal conductivity instrument Quickline-10 from ANTER Corporation, test conditions were set as follows: thickness of sample piece was 5 mm, diameter of sample piece was 50 mm, and test temperature was set at 43 °C.

An acceptable thermal conductivity value of the EMC for full-molded modules is at least 2.0 W/M·K, and the higher the better.

Water absorption:

- 15 Water absorption value was determined in accordance with the method of "PCT 24". Sample Piece size was set as $\Phi 50 \times 3$ mm; and test condition was 121 °C/ 100RH%/ 2atm/ 24hrs. Water absorption value can be calculated as Weight increment of sample piece after PCT24hrs / Weight of sample piece*100%.

- 20 An acceptable water absorption value of the EMC is less than 1.0%, preferably less than 0.8%, more preferably less than 0.6%.

Coefficient of thermal expansion (CTE1&2) and Glass transition temperature (Tg):

- 25 The CTE1&2 and Tg values of the molded composition were determined using a thermomechanical analyzer Q-400 from TA Instruments, and test conditions were set as follows: the sample piece was set as $\Phi 5 \times (5 \pm 0.1)$ mm, the sample piece was heated from room temperature to 280 °C at a rate of 10 °C/min, and the load was 0.1 N. CTE1 was calculated in a temperature range of 50 °C to 70 °C, CTE2 was calculated in a temperature range of 220 °C to 240 °C, and Tg was obtained in a temperature of 100 °C to 220 °C.

An acceptable CTE1 value is in a range of from 6 ppm to 45 ppm, preferably from 10 ppm to 40 ppm. An acceptable CTE2 value is in a range of from 20 ppm to 90 ppm, preferably from 25 ppm to 80 ppm.

5 An acceptable Tg value of the EMC is in a range of from 90 °C to 210 °C, but the higher the better. Preferably, the Tg value is higher than 130 °C, more preferably, higher than 145 °C.

Wire sweep:

10 The wire sweep value was determined by the following method: putting the encapsulated sample onto an X-ray emitter by which to obtain a first soldered dot projection and a second soldered dot projection of the wire; connecting the two dot-projections so as to obtain a base line with a projection length of L; picking the highest point of the camber line caused by wire sweep and making a tangent line at the highest point in parallel with the base line; measuring the distance A from the tangent line to the base line; calculating the wire sweep value by the formula of
15 $A/L * 100\%$.

An acceptable wire sweep value is less than 10%, and the lower the better.

Table 1: (in wt. %)

Components		Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5
Epoxy resin	DCPD ¹⁾	9.1	9.8	6	8.6	5
	EOCN ²⁾				1	
	BP ³⁾			2.5		2.5
	Bis-AN ⁴⁾					
Phenolic resin	PN ⁵⁾	3.2	4.1	3.5	3.4	1.5
	MFP ⁶⁾					
	Xylok resin ⁷⁾					2.5
Curing accelerator	TPP ⁸⁾			0.1	0.1	
	2P4MZ ⁹⁾	0.45	0.25	0.25		0.1
	2MZ ¹⁰⁾		0.2			
	DBU ¹¹⁾				0.2	0.2
silica	Crystalline silica ¹²⁾	76.3	76	77	75	77
	Spherical silica ¹³⁾	4	3	3	4.5	3.5
Flame retardant	Zinc borate ¹⁴⁾	4.5	4	5.5	5	5
	Aluminum hydroxide ¹⁵⁾					
Coupling agent	Epoxy silane-type ¹⁶⁾	0.45	0.3	0.3		
	Aminosilane-type ¹⁷⁾	0.3	0.55	0.45	0.5	0.5
	Mercaptosilane-type ¹⁸⁾				0.25	0.35
Wax	Aliphatic ester wax ¹⁹⁾	0.3	0.3	0.3	0.3	0.3
	Polyethylene wax ²⁰⁾	0.3	0.3	0.6	0.3	0.3
	Fatty acid ester wax ²¹⁾	0.3	0.4		0.4	0.4
Pigment	Carbon black ²²⁾	0.3	0.3	0.3	0.25	0.35
Ion trapping agent	Magnesium Aluminum hydroxide type ²³⁾	0.2	0.2	0.2	0.2	0.2
Stress absorbing agent	Polybutadiene-type ²⁴⁾	0.3	0.3			0.3
Total		100	100	100	100	100
Gel Time (s)		34	33	34	30	35
Spiral Flow (inch)		33	26	37	30	42
Viscosity (Pa·S)		25.3	21.4	23.5	24.2	18.9
Thermal Conductivity (W/M·K)		2.01	2.12	2.05	2.11	2.35
Water Absorption (%)		0.38	0.40	0.35	0.34	0.41
Tg (°C)		184	164	160	176	162
CTE1 (ppm)		19	20	19	20	19
CTE2 (ppm)		51	65	63	62	58
Wire sweep (%)		6.5	6.7	6.9	5.4	5.7

Table 1: (continued-1) (in wt. %)

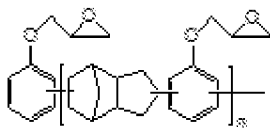
Components		Ex. 6	Ex. 7	Ex. 8	Ex. 9	Ex. 10
Epoxy resin	DCPD ¹⁾	4.5	8	4.3	5.5	9
	EOCN ²⁾	3	2	6.4		1
	BP ³⁾					
	Bis-AN ⁴⁾				2	
Phenolic resin	PN ⁵⁾	1.5	3.7	4.0	1.6	4.1
	MFP ⁶⁾	1.5				
	Xylok resin ⁷⁾				2.5	
Curing accelerator	TPP ⁸⁾	0.15		0.2		
	2P4MZ ⁹⁾	0.2	0.35		0.2	0.2
	2MZ ¹⁰⁾				0.2	0.15
	DBU ¹¹⁾			0.1		
silica	Crystalline silica ¹²⁾	78	73	74	79.9	77.3
	Spherical silica ¹³⁾	3.5	5	4		
Flame retardant	Zinc borate ¹⁴⁾	5	2.5	4	3	
	Aluminum hydroxide ¹⁵⁾		2.5		2.5	5
Coupling agent	Epoxy silane-type ¹⁶⁾	0.25	0.25	0.25	0.53	0.4
	Aminosilane-type ¹⁷⁾	0.3	0.5	0.5		0.3
	Mercaptosilane-type ¹⁸⁾	0.25	0.25	0.25	0.3	0.3
Wax	Aliphatic ester wax ¹⁹⁾	0.3	0.3	0.3	0.3	0.3
	Polyethylene wax ²⁰⁾	0.3	0.3	0.3	0.22	0.3
	Fatty acid ester wax ²¹⁾	0.4	0.4	0.4	0.4	0.4
Pigment	Carbon black ²²⁾	0.35	0.35	0.35	0.35	0.35
Ion trapping agent	Magnesium Aluminum hydroxide type ²³⁾	0.2	0.2	0.2	0.2	0.4
Stress absorbing agent	Polybutadiene-type ²⁴⁾	0.3	0.4	0.45	0.3	0.5
Total		100	100	100	100	100
Gel Time (s)		35	33	28	36	32
Spiral Flow (inch)		46	34	29	44	34
Viscosity (Pa·S)		20.4	18.3	33.4	16.1	16.8
Thermal Conductivity (W/M·K)		2.10	2.13	2.00	2.02	2.05
Water Absorption (%)		0.39	0.42	0.45	0.36	0.31
Tg (°C)		170	158	163	161	181
CTE1 (ppm)		21	21	22	20	20
CTE2 (ppm)		63	59	61	60	62
Wire sweep (%)		6.3	3.4	6.7	7.0	5.9

Table 1: (continued-2) (in wt. %)

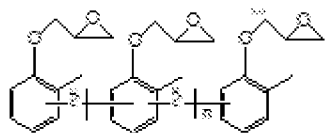
Components		Com. Ex. 1	Com. Ex. 2
Epoxy resin	DCPD ¹⁾		
	EOCN ²⁾	10	9.1
	BP ³⁾		
	Bis-AN ⁴⁾		
Phenolic resin	PN ⁵⁾	5.1	3.8
	MFP ⁶⁾		
	Xylok resin ⁷⁾		2
Curing accelerator	TPP ⁸⁾	0.2	0.3
	2P4MZ ⁹⁾		
	2MZ ¹⁰⁾		
	DBU ¹¹⁾	0.15	0.1
silica	Fused silica ²⁵⁾	8	3
	Spherical silica ¹³⁾	70	75
Flame retardant	Zinc borate ¹⁴⁾	5	5
	Aluminum hydroxide ¹⁵⁾		
Coupling agent	Epoxy silane-type ¹⁶⁾	0.4	0.3
	Aminosilane-type ¹⁷⁾	0.3	0.5
	Mercaptosilane-type ¹⁸⁾		
Wax	Aliphatic ester wax ¹⁹⁾	0.3	0.3
	Polyethylene wax ²⁰⁾	0.2	0.3
	Fatty acid ester wax ²¹⁾		
Pigment	Carbon black ²²⁾	0.35	0.3
Ion trapping agent	Magnesium Aluminum hydroxide type ²³⁾		
Stress absorbing agent	Polybutadiene-type ²⁴⁾		
Total		100	100
Gel Time (s)		30	28
Spiral Flow (inch)		33	34
Viscosity (Pa·S)		27.8	32.6
Thermal Conductivity (W/M·K)		0.73	0.65
Water Absorption (%)		0.41	0.45
Tg (°C)		174	173
CTE1 (ppm)		20	22
CTE2 (ppm)		59	64
Wire sweep (%)		7.8	8.2

Notes:

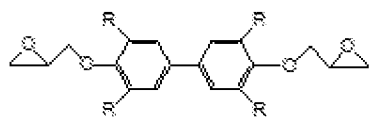
1): Dicyclopentadienephenol epoxy resin (DCPD), which has the following general structure:



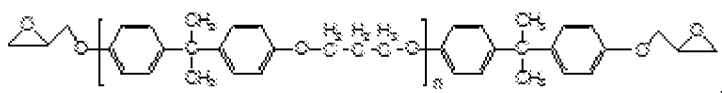
2): O-cresol novolac epoxy resins (EOCN), which has the following general structure:



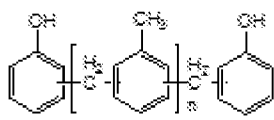
3): Biphenyl epoxy resin (BP), which has the following general structure:



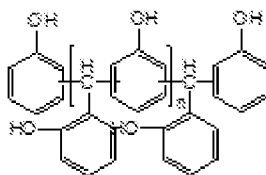
4): Bisphenol A novolac epoxy resin (Bis-AN), which has the following general structure:



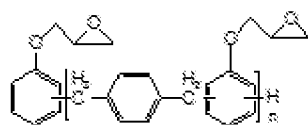
5): Phenol novolac resin (PN), which has the following general structure:



6): Multi-functional phenolic resin (MFN), which has the following general structure:



7): Xylok resin, which has the following general structure:



8): Triphenyl phosphine (TPP),

9): 2-phenyl-4-methyl imidazole (2P4MZ),

10): 2,4-diamino-6[2'-methylimidazolyl-(1'')]ethyl-s-triazine (2MZ),

11): 1,8-diazabicyclo(5,4,0) undecene-7 (DBU),

12): Crystalline silica, which has a particle size distribution of 15-45% in the range of

0.1-10 μ m, and 55-85% in the range of 10-75 μ m, and has an average particle size of 20-50 μ m,

13): Spherical silica, which has a particle size in the range of 0.1-30 μ m and an average particle size of 0.2-25 μ m,

14): Zinc borate,

15): Aluminum hydroxide,

16): Epoxidized silicone glycidyl resin,

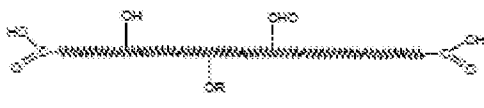
17): Phenylamino propyltrimethoxysilane,

18): 3-Mercaptopropyltrimethoxysilane,

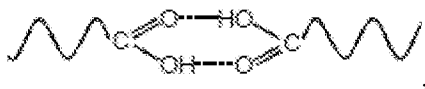
19): Aliphatic ester wax, which has the following general structure:



20): Polyethylene wax, which has the following general structure:



21): Fatty acid ester wax, which has the following general structure:



22): Carbon black,

23): Magnesium Aluminum hydroxide hydrate,

24): Polybutadiene-type stress absorbing agent,

25): Fused silica, which has a particle size in the range of 0.1-30 μ m and an average particle size of 0.2-25 μ m.

It can be seen from Table 1 that all epoxy molding compounds of Examples 1-10 of the present invention have a thermal conductivity of at least 2.0 W/M·K while the epoxy molding compounds of comparative examples 1-2 have a thermal conductivity of less than 2.0 W/M·K. Furthermore, all epoxy molding compounds of Examples 1-10 of the present invention have a low wire sweep of less than 7%. Meanwhile,

other properties as required for the EMC of all epoxy molding compounds of Examples 1-10 of the present invention are well-maintained.

The present invention is illustrated in details in the embodiments. However, it is apparent for those skilled in the art to modify and change the embodiments without
5 deviating from the spirit of the invention. All the modifications and changes should fall in the scope of the appended claims of the present application.

CLAIMS:

1. An epoxy molding compound comprising:

- (a) an epoxy resin,
- (b) a phenolic resin,
- (c) a curing accelerator,
- (d) crystalline silica, optionally in combination with spherical silica, and
- (e) one or more additives selected from the group consisting of a flame retardant, a wax, a coupling agent, a pigment, an ion trapping agent and a stress absorbing agent;

wherein the particle size distribution of the crystalline silica is that 5-50 wt.%, preferably 5-45 wt.% of the crystalline silica is less than 10 μm and 50-95 wt.%, preferably 55-95 wt.% of the crystalline silica is greater than 10 μm but less than 75 μm ,

the spherical silica has a particle diameter in the range of from 0.1 to 30 μm , preferably from 0.1 to 20 μm , more preferably 0.1 to 10 μm ,

the total amount of the crystalline silica and the spherical silica (if present) is about 40-90wt.%, preferably about 50-88wt.%, more preferably about 60-85 wt.% based on the total weight of the composition,

the weight ratio between the crystalline silica and the spherical silica (if present) is in a range of from 5:1 to 35:1, preferably from 10:1 to 30:1, more preferably from 12:1 to 28:1, and

the molar ratio between the hydroxyl groups in the phenolic resin and the epoxy groups in the epoxy resin is in a range of from 0.5 to 2.0, preferably from 0.8 to 1.7, more preferably from 0.9 to 1.5.

2. The epoxy molding compound according to claim 1, wherein the epoxy resin is one or more selected from o-cresol novolac epoxy resins (EOCN), dicyclopentadienephenol epoxy resins (DCPD), biphenyl epoxy resins, bisphenol A novolac epoxy resins, triphenylmethane epoxy resins, epoxy resins having a naphthalene skeleton, multi-aromatic epoxy resins (MAR) and multi-functional epoxy resins (MFN).

3. The epoxy molding compound according to claim 1 or 2, wherein the phenolic resin is one or more selected from phenol novolac resins, Xylok resins, multi-aromatic phenolic resins and multi-functional phenolic resins.
4. The epoxy molding compound according to any one of claims 1 to 3, wherein the curing accelerator is one or more selected from triphenyl phosphine (TPP), 1,8-diazabicyclo(5,4,0) undecene-7 (DBU), 2,4-diamino-6[2'-methylimidazolyl-(1')]ethyl-s-triazine (2MZ), 2-phenyl-4-methyl imidazole (2P4MZ), N,N-dimethyl benzyl amine and triethylamine.
5. The epoxy molding compound according to any one of claims 1 to 4, wherein the flame retardant is one or more selected from zinc borate, aluminum hydroxide, magnesium hydroxide, titanium oxide, calcium silicate, brominated epoxy resin and antimony trioxide.
6. The epoxy molding compound according to any one of claims 1 to 5, wherein the wax is one or more selected from montan waxes, fatty acid ester waxes, fatty acid waxes, aliphatic ester waxes, polyethylene waxes, polypropylene waxes, alkyl oligomer waxes and amide waxes.
7. The epoxy molding compound according to any one of claims 1 to 6, wherein the coupling agent is one or more selected from epoxysilanes, aminosilanes, mercaptosilanes, ureidosilanes, vinylsilanes and alkylsilanes..
8. The epoxy molding compound according to any one of claims 1 to 7, wherein the ion trapping agent is one or more selected from magnesium aluminum hydroxide type, aluminosilicate, antimony hydroxide type and bismuth hydroxide type .
9. The epoxy molding compound according to any one of claims 1 to 8, wherein the stress absorbing agent is one or more selected from silicone oil, silicone resins, and butadiene-type rubbers.
10. The epoxy molding compound according to any one of claims 1 to 9, wherein the compound comprises, based on the total weight of the epoxy compound:

- (a) 1-25 wt.% of the epoxy resin,
- (b) 1-20 wt.% of the phenolic resin,
- (c) 0.01-5 wt.% of the curing accelerator,
- (d) 40-90 wt.% of the crystalline silica and the spherical silica (if present), and
- (e) 0.1-45 wt.% of one or more additives.

11. The epoxy molding compound according to any one of claims 1 to 9, wherein the compound comprises, based on the total weight of the epoxy compound:

- (a) 5-20 wt.% of the epoxy resin,
- (b) 2-15 wt.% of the phenolic resin,
- (c) 0.01-2 wt.% of the curing accelerator,
- (d) 50-88 wt.% of the crystalline silica and the spherical silica (if present), and
- (e) 0.5-40 wt.% of one or more additives.

12. A method for preparing the epoxy molding compound of any one of claims 1 to 11, comprising steps of:

- (1) weighing up all of the components and mixing them in a high speed mixer to get a premixed powder; and
- (2) extruding the premixed powder by an extruder to get an extrusion product and then crushing the extrusion product into powder.

13. The method according to claim 12, wherein the extruder is a twin-screw extruder.

14. The method according to claim 12 to 13, wherein the extrusion is performed at 60~110°C with a rotation speed of 50~250 rpm.

15. Use of the epoxy molding compound according to any one of claims 1 to 11 for full-molded modules.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2015/074575

A. CLASSIFICATION OF SUBJECT MATTER

C08L 63/00(2006.01)i; C08K 13/02(2006.01)i; C08K 3/36(2006.01)i; C08K 7/00(2006.01)i; C08K 7/18(2006.01)i

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)

C08L63/-; C08K13/-; C08K3/-; C08K7/-; C08K5/-

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)

CNKI, CANPAT, WPI, EPODOC: epoxy resin, molding, phenolic resin, crystalline silica, spherical silica, flame, wax, coupling agent, pigment, ion trapping agent, stress absorbing agent, particle size, diameter, catalyst, filler

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CN 103421275 A (HENKEL HUAWEI ELECTRONICS CO. LTD.) 04 December 2013 (2013-12-04) description, paragraphs [0004]-[0015] and example 1	1-15
A	CN 1605599 A (SUN, ZHONGXIAN) 13 April 2005 (2005-04-13) claims 1-4, description, page 1 line 29 to page 2 line 21	1-15
A	CN 103450632 A (HENKEL HUAWEI ELECTRONICS CO. LTD.) 18 December 2013 (2013-12-18) claims 1-11, description, paragraphs [0006]-[0008]	1-15
A	CN 101544841 A (GUANGDONG SHENGYI SCI. TECH. CO.) 30 September 2009 (2009-09-30) claims 1, 5-7, description, page 4, lines 12-18	1-15
A	CN 103421279 A (HENKEL HUAWEI ELECTRONICS CO. LTD.) 04 December 2013 (2013-12-04) claims 1-13, description, paragraphs [0006]-[0009] and examples	1-15



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

“E” earlier application or patent but published on or after the international filing date

“L” document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

“O” document referring to an oral disclosure, use, exhibition or other means

“P” document published prior to the international filing date but later than the priority date claimed

“T” later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

“X” document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

“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

13 October 2015

Date of mailing of the international search report

25 November 2015

Name and mailing address of the ISA/CN

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PEI,Shaoping

Telephone No. (86-10)82246781

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2015/074575

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CN 103421272 A (HENKEL HUAWEI ELECTRONICS CO. LTD.) 04 December 2013 (2013-12-04) claims 1-11, description, paragraphs [0006], [0007] and examples	1-15

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.

PCT/CN2015/074575

Patent document cited in search report			Publication date (day/month/year)	Patent family member(s)			Publication date (day/month/year)
CN	103421275	A	04 December 2013	None			
CN	1605599	A	13 April 2005	None			
CN	103450632	A	18 December 2013	None			
CN	101544841	A	30 September 2009	US	8269115	B2	18 September 2012
				AU	2010200479	B2	13 October 2011
				US	2010258340	A1	14 October 2010
				EP	2241589	A3	18 January 2012
				AU	2010200479	A1	28 October 2010
				EP	2241589	A2	20 October 2010
				CN	101544841	B	21 July 2010
CN	103421279	A	04 December 2013	None			
CN	103421272	A	04 December 2013	None			