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(54) **Title:** EPOXY RESIN COMPOSITION

(57) **Abstract:** The present invention relates to an epoxy resin composition, in particular, to an epoxy resin composition suitable for use as an encapsulation agent for electrical parts such as drive- ICs mounted on a circuit board. The epoxy composition is a liquid phase at room temperature and includes 100 parts by weight of an epoxy resin having 2 or more glycidyl groups in the molecure, 10-50 parts by weight of a reactive diluent having one or more glycidyl groups in the molecure, 50-200 parts by weight of an anhydrous curing agent which is a liquid phase at room temperature with respect to 100 parts by weight of the epoxy resin and the reactive diluent, and 1-20 parts by weight of a latent curing accelerator with respect to 100 parts by weight of the epoxy resin, the reactive diluent, and the anhydrous curing agent, and whose viscosity at 25 °C is 250-500cps, gelation time at 150°C is below 120 seconds.

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[DESCRIPTION]

[Invention Title]

EPOXY RESIN COMPOSITION

[Technical Field]

The present invention relates to an epoxy resin composition, in particular, to an epoxy resin composition suitable for use as an encapsulant for electrical parts such as driver-ICs mounted on a circuit board.

[Background Art]

Recently, in response to the trends of miniaturized, light-weighted and slim electrical/electronic devices such as cellular phones, notebook computers and wall hanging TVs, flat panel displays such as LCD display, plasma display, electroluminescent(EL) device have been mainly used for image display devices, and the miniaturization of driver-IC(Integrated Circuit) that drives these electric components is required. Serial steps to mount bare chips like driver-IC on a circuit board are referred to as packaging. As for the packaging method, the tape carrier package (TCP) process which includes forming circuits on a polyimide film, mounting a bare chip, turning the bare chip downward and injecting a resin into a hole of the circuit board, was used

previously.

[Disclosure]

[Technical Problem]

However, the TCP process has shortcomings that the package components have low flexibility and has limitation to reduce weight, thickness, length and size of the package components, because after processing a circuit board where a thick polyimide film and copper foil are attached by an epoxy-based adhesive, the chip was mounted on the circuit board.

To address this problem, a COF(Chip On Film) underfill process that includes forming an insulating film having superior flexibility by coating a thin polyimide on the upper portion of a thin copper foil or sputtering copper layer on the surface of thin polyimide film, producing a flexible circuit board by forming a lead wiring on the insulating film, mounting a bare chip on the flexible circuit board, filling the gap between the board and the bare chip with resin has been developed and is used recently. In such a COF process, since the gap between the board and the bare chip is less than several tens of micrometers, there is a problem that the existing resin for TCP process including a large amount of inorganic fillers and solvents can not be directly applied to the COF underfill process. Further, because the gap between the board and the bare chip is narrowed down to 10 um and the size of the chip is getting larger, needs for

encapsulation composition which has better fillability and is uniformly cured is on the rise.

[Technical solution]

Therefore, the present invention provides an epoxy resin composition which has better penetrability and underfill ability into the gap between a board and a bare chip, and which can be uniformly cured over the whole surface area of the chip. The present invention further provides an epoxy resin composition having superior productivity achieved by short thermosetting time, and excellent workability achieved by forming less bubbles and volatile components. The invention further provides an epoxy resin composition, not only having superior fillet forming ability and adhesion so that it firmly connects a circuit board and a bare chip, but also having superior temperature cycling ability and properties under high temperature and high humidity, so that it can produce chip packages having high reliability.

To achieve the objectives, the present invention provides an epoxy composition which is a liquid phase at room temperature and includes 100 parts by weight of an epoxy resin having 2 or more glycidyl groups in the molecule, 10-50 parts by weight of a reactive diluent having one or more glycidyl groups in the molecule, 50-200 parts by weight of an anhydride curing agent which is a liquid phase at room temperature with respect to 100 parts by weight of the epoxy resin and the reactive diluent, and 1-20 parts

by weight of a latent curing accelerator with respect to 100 parts by weight of the epoxy resin, the reactive diluent, and the anhydrous curing agent, and whose viscosity at 25℃ is 250-500cps, gelation time at 150℃ is 120 seconds or below.

Hereinafter, more detailed descriptions will be given.

The inventors completed this invention by controlling the gelation time and viscosity of a composition by adjusting components and contents of an epoxy resin composition as a result of broad researches for an epoxy resin composition which is useful for an underfill composition of the COF process. Main components of the epoxy resin composition according to the invention include an epoxy resin, a reactive diluent, an anhydrous curing agent and a latent curing accelerator.

The epoxy resin used in the invention, which is liquid at room temperature, has 2 or more glycidyl groups in a molecule, and a variety of curable epoxy resins may be used, for example, bisphenol A type epoxy resin, bisphenol F type epoxy resin may be used individually or together with. Specific examples of the bisphenol A type(diglycidyl ether) epoxy resin include EpikoteYL-980(JAPAN EPOXY RESIN CO. LTD.), RE-310(NIPPON KAYAKU CO. LTD.), DER-332(The Dow Chemical Company), etc., and specific examples of the bisphenol F type(diglycidyl ether) epoxy resin include Epikote YL-983U(JAPAN EPOXY RESIN CO. LTD.), RE-304(NIPPON KAYAKU CO. LTD.), RE-404(NIPPON KAYAKU CO. LTD.), RE-303S(NIPPON KAYAKU CO. LTD.), etc.

The reactive diluent used in the invention functions to decrease viscosity of the

composition, a variety of compounds having one or more glycidyl groups in the molecule may be used, for example, alkyl monoglycidyl ether, butyl phenyl glycidyl ether, alkyl phenol monoglycidyl ether, polyglycol diglycidyl ether, alkyl diglycidyl ether, etc. may be used individually or together with. Here, the carbon number of the alkyl group is preferably 1-10, more preferably 1-4. Specific examples of mono-functional reactive diluent having one glycidyl group in the molecule include YED-111E(JAPAN EPOXY RESIN CO. LTD.), YED-122(JAPAN EPOXY RESIN CO. LTD.), ED-501(ADEKA CORPORATION), ED-502(ADEKA CORPORATION), ED-509S(ADEKA CORPORATION), ED-529(ADEKA CORPORATION), ED-518(ADEKA CORPORATION), etc., specific examples of di-functional reactive diluent having two glycidyl groups in the molecule include YED-216(JAPAN EPOXY RESIN CO. LTD.), ED-503(ADEKA CORPORATION), ED-506(ADEKA CORPORATION), ED-523T(ADEKA CORPORATION), ED-515(ADEKA CORPORATION), etc., specific examples of multi-functional reactive diluent include YED-205(JAPAN EPOXY RESIN CO. LTD.), ED-505R(ADEKA CORPORATION), etc. The amount of the reactive diluent is preferably added by 10-50 parts by weight with respect to 100 pars by weight of the epoxy resin, more preferably 20-40 parts by weight. If the reactive diluent is added by less than 10 parts by weight, viscosity of the composition may not decrease sufficiently, if more than 50 parts by weight, glass transition temperature(Tg) and mechanical property of the composition may be deteriorated.

The anhydride curing agent is what reacts with the epoxy resin and cures, so that the viscosity of the epoxy resin composition becomes 250-500cps, it is preferable that a liquid phase anhydride curing agent be used, among them, an anhydride curing agent that generates less carbon dioxide during the curing, for example, an anhydride curing agent produced by reaction of triene or diene of C_{10} with anhydride maleic acid, is preferable. Specific examples of the anhydride curing agent are 3,4-dimethyl-6-(2-methyl-1-prophenyl)-4-cyclohexene-1,2-dicarboxylic acid anhydride or its mixture with 1-isopropyl-4-methylbicyclo[2.2.2]octa-5-ene-2,3-dicarboxylic anhydride. YH-306(JAPAN EPOXY RESIN CO. LTD.), YH-307(JAPAN EPOXY RESIN CO. LTD.) is commercially available. Appropriate adding amount of the anhydride curing agent is 50-200 parts by weight with respect to 100 parts by weight of the epoxy resin and the reactive diluent, preferably, 100-150 parts by weight. If the anhydride curing agent is added by less than 50 parts by weight or more than 200 parts by weight, unreacted epoxy resin or unreacted anhydrous curing agent remains behind and can cause deterioration of glass transition temperature(Tg) and mechanical property.

The latent curing accelerator used in the invention may be preserved in a mixture with the epoxy resin and the anhydride curing agent and function to cure the composition when the reaction of the anhydride curing agent is initiated by means of heating and so on. For example, micro-encapsulated amine compounds, imidazole compounds and modified amine compounds and modified imidazole compounds may

be used. Examples of commercialized products of the latent curing accelerator are MY-24, MY-H, MY-D(the foregoing available from AJINOMOTO-FINE-TECHNO CO.,INC.), Fujicure FXR-1020, FXR-1030, FXR-1080, FXR-1110(the foregoing available from FUJI KASEI CO., INC.), novacure HX-3088, HX-3722, HX3748, HX-3921, HX-3941HP(the foregoing available from ASAHI KASEI CHEMICALS CORPORATION). The adding amount of the latent curing accelerator is 1-20 parts by weight, preferably 2-10 parts by weight with respect to 100 parts by weight of the anhydrous curing agent. If the curing accelerator is added by less than 1 part by weight, the curing rate increasing effect is low. On the other hand, if it is added by more than 20 parts by weight, viscosity of the composition increases and the mechanical property may be deteriorated. The liquid epoxy resin composition for encapsulation according to the invention has a low viscosity of 250-500cps at 25℃, and gelation time of below 120 seconds at 150℃, preferably 80-120 seconds. Any general inorganic filler, coupling agent, antifoaming agent, pigment, dye, curing accelerator, etc may be added within a scope apparent to those skilled in the art. The liquid epoxy resin composition according to the invention may be produced by sufficiently mixing each compound by generally known methods, for example, uniformly mixing each compound using a spiral mixer, a universal mixer, etc. under a vacuum condition. A thermosetting one-pack type epoxy resin composition according to the invention, for example, in case of producing a semiconductor device by mounting a bare chip such as flip chip on a flexible circuit

board of polyimide by COF packaging method, is particularly useful for an underfill composition for the flip chip.

[Mode for Invention]

Hereinafter, explanations will be given in greater detail with specific examples. The invention is not limited to the examples stated below and it is also apparent that more changes may be made by those skilled in the art without departing from the principles and spirit of the present invention.

[Example 1]

70 parts by weight of a bisphenol F type epoxy resin (JAPAN EPOXY RESIN CO. LTD., Product name: Epikote YL983U), 30 parts by weight of a mono-functional reactive diluent (JAPAN EPOXY RESIN CO. LTD., Product name: YED-111E), 120 parts by weight of a liquid anhydrous curing agent (JAPAN EPOXY RESIN CO. LTD., Product name: YH-306), 5 parts by weight of a curing accelerator (AJINOMOTO FINE TECHNO CO. LTD., Product name: MY-24) and 2.0 parts by weight of a silane coupling agent (JAPAN UNIKASA, Product name: A-187) were mixed to produce a liquid resin composition for encapsulation.

[Example 2]

70 parts by weight of a bisphenol F type epoxy(NIPPON KAYAKU CO. LTD.,

Product name: RE-303S), 30 parts by weight of a multi-functional reactive diluent(ASAHI DENKA CO. LTD., Product name: ED-505R), 130 parts by weight of a liquid anhydrous curing agent(JAPAN EPOXY RESIN CO. LTD., Product name: YH-307), 2.0 parts by weight of a silane coupling agent (GE TOSHIBA SILICONES, Product name: A-187), and 5 parts by weight of a curing accelerator (FUJI KASEI CO. LTD, Product name: FXR-1020) were mixed to produce a liquid resin composition for encapsulation.

[Comparison Example 1]

100 parts by weight of a bisphenol F type epoxy resin (JAPAN EPOXY RESIN CO. LTD., Product name: Epikote YL983U), 90 parts by weight of a liquid anhydrous curing agent (JAPAN EPOXY RESIN CO. LTD., Product name: YH-306), 0.5 parts by weight of a curing accelerator (FUJIKASEI CO. LTD, Product name: FXR-1040) and 2.0 parts by weight of a silane coupling agent (GE TOSHIBA SILICONES, Product name: A-187) were mixed to produce a liquid resin composition for encapsulation.

[Comparison Example 2]

70 parts by weight of a bisphenol F type epoxy(NIPPON KAYAKU CO. LTD., Product name: RE-303S), 30 parts by weight of a multi-functional reactive diluent(ADEKA CORPORATION, Product name: ED-505R), 210 parts by weight of a liquid anhydrous curing agent(JAPAN EPOXY RESIN CO. LTD., Product name: YH-307), 2.0 parts by weight of a silane coupling agent (GE TOSHIBA SILICONE, Product

name: A-187) and 0.9 parts by weight of a curing accelerator (AJINOMOTO FINE TECHNO CO. LTD., Product name: MY-H) were mixed to produce a liquid resin composition for encapsulation.

The viscosity of the one-pack type liquid epoxy resin composition for encapsulation, which were produced according to examples 1 and 2, and comparison examples 1 and 2, was determined at 25℃ with Haake cone and plate type viscometer, the gelation time at 150℃ was determined by dropping one drop of the composition on a micro-slide glass, stirring the composition with a wood stick, and monitoring the time when it has become impossible to stir the composition with melting point measuring apparatus from FISHER SCIENTIFIC CO. LTD., and decrease of weight for 15 minutes were estimated with TGA(Thermogravimetric analysis). The results are shown in table 1. Also, 2 glass plates of 2x2cm size in which a spacer was incorporated were placed on a heat plate of 130℃ so that a 20 micron size gap is maintained, then the fillability of the resin was evaluated by dropping the resin composition on one glass plate and estimating the time for filling to the opposite side glass plate. If the gelation time was below 10 seconds, it was decided as 'good' and if more than 10 seconds, 'defective', and the results are shown in Table 1.

Table 1.

	Example 1	Example 2	Comparison Example 1	Comparison Example 2
Viscosity	340cps	320cps	580cps	300cps
Gelation time	85 sec.	90 sec.	210 sec.	200 sec.
Decrease of weight	2.1%	2.3%	5.2%	4.5%
Fillability	Good	Good	Defective	Good

Referring to Table 1, it is shown that the liquid epoxy resin composition for encapsulation of the examples have superior resin viscosity and gelation time in comparison with epoxy resin composition of the comparison examples. Also, when the gelation time is longer, it is shown that the decrease of weight is getting higher because of evaporation of the liquid anhydride that occurs before the epoxy resin cures by the reaction, in this case, it is highly expected that bubbles are generated between chips and boards. Further, it is shown that in case of the viscosity is more than 500cps, it takes a long period of time to fill the gap between chips and boards with the epoxy resin, or incomplete filling of the gap occurs.

[Industrial Applicability]

As previously mentioned, the epoxy resin composition according to the

invention has better penetrability and underfill ability into the gap between a film and a bare chip, and can be uniformly cured over the whole surface area of the chip. Further, the epoxy resin composition has superior productivity achieved by short thermosetting time, excellent workability achieved by forming less bubbles and volatile components, and advantage to produce chip packages having high reliability.

[CLAIMS]

[Claim 1]

An epoxy resin composition comprising :

100 parts by weight of an epoxy resin which is a liquid phase at room temperature and has 2 or more glycidyl groups in a molecule;

10-50 parts by weight of a reactive diluent having one or more glycidyl groups in a molecule;

50-200 parts by weight of an anhydride curing agent which is a liquid phase at room temperature with respect to 100 parts by weight of the epoxy resin and the reactive diluent; and

1-20 parts by weight of a latent curing accelerator with respect to 100 parts by weight of the epoxy resin, the reactive diluent, and the anhydride curing agent,

wherein a viscosity of the epoxy composition at 25℃ is 250-500cps and gelation time at 150℃ is 120 seconds or below.

[Claim 2]

The epoxy resin composition of claim 1, wherein the epoxy resin is selected from the group consisting of a bisphenol A type epoxy resin, a bisphenol F type epoxy resin and mixtures thereof.

[Claim 3]

The epoxy resin composition of claim 1, wherein the reactive diluent has one glycidyl goup in a molecure.

[Claim 4]

The epoxy resin composition of claim 1, wherein the anhydride curing agent is produced by reaction of a triene or a diene with an anhydrous maleic acid.

[Claim 5]

The epoxy resin composition of claim 1, wherein the latent curing accelerator is micro-capsulated compound that is selected from the group consisting of amine compounds, imidazole compounds, modified amine compounds and modified imidazole compounds.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2006/005676

A. CLASSIFICATION OF SUBJECT MATTER

C08L 63/02(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)

IPC8: C08L 63/02

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

Electronic data base consulted during the intertnational search (name of data base and, where practicable, search terms used)

eKIPASS "underfill, epoxy, reactive diluent"

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 6,342,577 B1 (LOCTITE CORPORATION) 29 Jan. 2002 See abstract and claims 1-16.	1-5
A	US 2002/0106515 A1 (NEIL M. CARPENTER, MICHEL RUYTERS) 8 Aug. 2002 See abstract and claims 1-6.	1-5
A	US 6,180,696 B1 (GEORGIA TECH RESEARCH CORPORATION) 30 Jan. 2001 See abstract and claims 1-4.	1-5
A	US 2002/0142167 A1 (UBE INDUSTRIES, LTD.) 3 Oct. 2002 See abstract and claims 1-7.	1-5

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

☒ See patent family annex.

* Special categories of cited documents:

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

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

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
06 APRIL 2007 (06.04.2007)	**06 APRIL 2007 (06.04.2007)**

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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