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(54) RESIN COMPOSITION FOR PRINTED WIRING BOARD, PREPREG, LAMINATE, AND PRINTED WIRING BOARD MADE WITH THE SAME

HARZZUSAMMENSETZUNG FÜR LEITERPLATTE UND DAMIT HERGESTELLTE PREPREGS, LAMINATE UND LEITERPLATTEN

COMPOSITION DE RESINE POUR CARTES IMPRIMEES, PREIMPREGNE, LAMINE, ET CARTE IMPRIMEE OBTENUE

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- **DATABASE WPI Week 200379 Derwent Publications Ltd., London, GB; AN 2003-853778 XP002417089 & JP 2003 292733 A 15 October 2003 (2003-10-15) -& US 2004/147658 A1 (ICHIRYU A; ICHIYANAGI A; KUWAKO F; MATSUSHIMA T; MIWA H; SATO T; YAMAZ) 29 July 2004 (2004-07-29)**
- **NAKAMURA Y ET AL: "EFFECT OF PARTICLE SHAPE, SIZE AND INTERFACIAL ADHESION ON THE FRACTURE STRENGTH OF SILICA-FILLED EPOXY RESIN" 1999, POLYMERS AND POLYMER COMPOSITES, RAPRA TECHNOLOGY, SHAWBURY, SHREWSBURY, GB, PAGE(S) 177-186 , XP000832156 ISSN: 0967-3911 * abstract * * page 182; figure 6 ***

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Description

FIELD OF INVENTION

[0001] The present invention relates to an epoxy resin composition for a printed wiring board used in manufacturing of an electric laminated board, a prepreg for a printed wiring board, as well as a laminated board for a printed wiring board, a printed wiring board and a laminated printed wiring board, using this prepreg, for use in an electronic device.

BACKGROUND OF THE INVENTION

[0002] Dicyandiamide (DICY) has been used for a long period of time as a curing agent of epoxy resins for printed wiring boards. However, a material superior in a long term insulation reliability (CAF resistance) and a material with a high decomposition temperature required for lead-free soldering have been demanded as a requirement along with higher density wiring in printed wiring boards in recent years, so that phenol type curing systems superior in these properties have been used as the curing agent.
[0003] However, phenol type curing agents have problems such that they cannot impregnate well into a glass base substrate and prepregs prepared therefrom are poor in appearance.
[0004] In regards to improvement of appearance in prepregs, it is described in Japan Patent Kokai H07-48586 and Japan Patent Kokai H07-68380 that impregnation into the prepreg is improved by reacting tetrabromobisphenol A with both a bisphenol A type epoxy resin and a novolac type epoxy resin in preparation of prepregs; and it is described in Japan Patent 3395845 that appearance of prepregs is improved by using a bisphenol novolac resin with a softening temperature of 60 °C to 90 °C.

DISCLOSURE OF THE INVENTION

[0005] The present inventors have reached the present invention, finding that, through using a specific silica filler in phenol-curing resin prepregs for laminated boards, are effectively improved appearance of prepregs has been attained.
[0006] It is well known heretofore that fillers (filling materials) are used for reducing coefficient of thermal expansion (decreasing α), increasing rigidity or reducing water adsorptivity in laminated boards. Specifically, as described in Japan Patent Kokai H06-216484, low water adsorptivity has been achieved by using a spherical inorganic filler with a small specific surface area (0.2 m^2/g to 2.0 m^2/g).
[0007] However, the present inventors have found that addition of a silica filler, having a particular shape, a prescribed range of average particle diameter and a prescribed range of specific surface area, to an epoxy resin system using a phenolic curing agent, is capable of improving appearance of prepregs, through increasing apparent resin viscosity to prevent resin sagging within a drying oven, as well as not impairing penetrativity into a reinforcing material, since viscosity of the resin itself does not increase locally, and is capable of improving appearance of prepregs.
[0008] The present invention has been achieved in view of the above problems. It is an object of the present invention to provide a prepreg having good appearance, in system using a phenolic curing agent, which is superior in heat resistance, as well as to provide a metal foil-clad laminated board using the above prepreg and to provide a printed wiring board using this metal foil-clad laminated board.

SUMMARY OF THE INVENTION

[0009] In order to achieve the purpose mentioned above, is provided according to the present invention, an epoxy resin composition for a printed wiring board, comprising an epoxy resin, a phenol novolac resin, a curing accelerator and a silica filler, characterized in that, as the silica filler, is used a silica filler which has a shape having at least two planes, and has an average particle diameter between 0.3 μm and 10 μm and a relative surface area between 8 m^2/g and 30 m^2/g.
[0010] In the epoxy resin composition for a printed wiring board according to this invention, it is preferred to use a silica filler having at least two planes in the shape, an average particle diameter between 0.3 μm and 10 μm and a relative surface area between 10 m^2/g and 20 m^2/g.
[0011] In the epoxy resin composition for a printed wiring board of the invention, said silica filler is preferably added in an amount of from 3% to 80% by weight per the solid content of the resin.
[0012] In the epoxy resin composition for a printed wiring board of the invention, said silica filler is used preferably having an electric conductivity of 15 μs or less.
[0013] In the epoxy resin composition for a printed wiring board of the invention, as said silica filler, is used preferably a silica filler which has been vitrified through melting at a temperature of 1800 °C or higher.
[0014] In another aspect of the epoxy resin composition for a printed wiring board of the present invention, is used an

epoxy resin which has a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler, and contains an epoxy resin obtainable by reacting a dihydric phenol with a bisphenol A type epoxy resin in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

[0015] In still another aspect of the epoxy resin composition for a printed wiring board of this invention, is used an epoxy resin having a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler and containing an epoxy resin possessing a dicyclopentadienyl structure in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

[0016] In yet another aspect of the epoxy resin composition for a printed wiring board of the invention, is used an epoxy resin having a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler and containing of a novolac type epoxy resin in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

[0017] In further aspect of the invention, the epoxy resin composition for a printed wiring board does not contain any bromine.

[0018] A prepreg according to the present invention is manufactured by impregnating a reinforced material with the epoxy resin composition for a printed wiring board as described above, followed by drying and semi-curing the composition to B-stage.

[0019] A laminated board according to this invention is prepared by gluing the prepreg as above to a metal foil surface and hot pressing them.

[0020] A printed wiring board according to the invention is prepared by using the metal foil clad laminated board mentioned above.

DISCLOSURE OF THE INVENTION IN DETAIL

[0021] Embodiments according to the present invention are described below.

[0022] It is essential in this invention that the epoxy resin composition for a printed wiring board used therein is comprised of an epoxy resin, a phenol novolac resin, a curing accelerator and a silica filler. The composition may comprise an organic solvent and others, such as a UV screener and a fluorescent agent as needed.

[0023] Epoxy resins are not particularly limited, but include, for example, bifunctional epoxy resins, such as bisphenol A type epoxy resins, bisphenol F type epoxy resins and tetrabromobisphenol A type epoxy resins, or brominated derivatives of these; novolac type epoxy resins, such as cresol novolac type epoxy resins or brominated derivatives thereof; dicyclpentadienyl type epoxy resins; biphenyl type epoxy resins, such as tetramethylbiphenyl epoxy resins; polyfunctional epoxy resins, such as a trifunctional epoxy resins and tetrafunctional epoxy resins; and hydroquinone type epoxy resins or brominated derivatives thereof. These resins can be used singly or in combination of two or more as a mixture.

[0024] Among brominated epoxy resins as mentioned above, preferred, in view of achieving well balanced fire retardancy of the cured product with cost, are ones having a bromine content of between 5% and 20% by weight per the solid content of the whole resin (without silica filler), and containing an epoxy resin obtainable by reacting a dihydric phenol with a bisphenol A type epoxy resin in an amount of at least 40% and up to 100% by weight based on the whole amount of the epoxy resin solid content.

[0025] Besides, among brominated epoxy resins as above, preferred, for the purpose of attaining lower hygroscopicity with retaining good fire retardancy of the cured product, are ones having a bromine content of between 5% and 20% by weight per the solid content of the whole resin (without silica filler), and containing an epoxy resin having a dicylopentadienyl structure in an amount of at least 40% and up to 100% by weight based on the whole amount of the epoxy resin solid content.

[0026] In addition, among brominated epoxy resins as above, preferred, in regard of providing higher glass transition temperature of the cured product with retaining fire retardancy, are ones having a bromine content of between 5% and 20% by weight per the solid content of the whole resin (without silica filler), and containing a novolac type epoxy resin in an amount of at least 40% and up to 100% by weight based on the whole amount of the epoxy resin solid content.

[0027] Moreover, ones comprising an epoxy resin free from any a bromine are halogen-free and preferred for low environmental load.

[0028] Phenol novolac resins, usable as the curing agent in this invention, are not particularly limited, but include, for example, phenol novolac resins, obtainable by reacting formaldehyde with phenols, such as phenol and cresol; and bisphenol novolac resins, obtainable by reacting formaldehyde with bisphenols, such as bisphenol A. These resins can be used singly or in combination of two or more.

[0029] Further, in case of mixing a phenol novolac resin curing agent as one component of the epoxy resin composition according to the present invention, it is preferred, for providing cured products having well balanced properties of glass transition temperature, peal strength and the like, to use an equivalent ratio of the epoxy group to the phenolic hydroxyl group in the range of 1:1.2 to 1:0.7.

[0030] As an organic solvent according to the invention, ketones, such as methyl ethyl ketone and cyclohexanone;

and cellosolves, such as methoxypropanol can be preferably used.

[0031] Curing accelerators according to the invention are not particularly limited, but include, for example, imidazoles, such as 2-methylimidazole, 2-ethyl-4-methyl- imidazole, 2-phenylimidazole and 1-cyanoethyl-2-ethylimidazole; tertiary amines, such as benzyldimethylamine; organic phosphines, such as tributylphosphine and triphenyl- phosphine, and imidazolesilanes. These compounds can be used singly or in combination of two or more.

[0032] A silica filler used in the invention has a shape possessing at least two planes, an average particle diameter at least 0.3 μm and up to 10 μm, and a specific surface area at least 8 m^2/g and at most 30 m^2/g, preferably at least 10m^2/g and at most 20 m^2/g.

[0033] The shape (geometry) of the above silica filler has at least two planes, and it is not spherical in shape. For example, there may be mentioned an indefinite shape obtainable by crushing.

[0034] Silica filler, possessing the same particle diameter as the above but have a specific surface area of 8 m^2/g or less, has little effects of increasing an apparent resin viscosity and has no effect of preventing resin sagging during drying. In addition, a silica possessing a specific surface area exceeding 30m^2/g is not commercially available as an industrial product so that it cannot be used.

[0035] It is preferred to added the above silica filler in an amount of at least 3% and at most 80% by weight per the whole resin solid content.

[0036] In case of adding the above silica filler in an amount below 3% by weight, no advantage may be realized by its addition in some cases. In case of added amount exceeding 80% by weight, problems, in peal strength and easiness in drilling process, might possibly be generated.

[0037] In addition, it is preferred that above silica filler has an electric conductivity of 15 μs/cm (microsiemens/cm) or less in order to retain reliability with electric insulation.

[0038] Further, among silica fillers as mentioned above, preferred are ones obtained by melting at 1800 °C or higher to a vitrified form, because it provides cured products of lower coefficient of thermal expansion. Furthermore, this vitrified product is preferred since it is capable of eliminating crystalline substances, which are hazardous substance.

[0039] In addition, the silica filler mentioned above is preferably surface-treated with a silane coupling agent or the like to improve the interfacial strength between the resin and the silica filler.

[0040] A prepreg for a printed wiring board according to the present invention can be manufactured, for instance, by preparing a varnish of an epoxy resin composition for printed wiring board as above with an organic solvent as mentioned previously, impregnating the varnish into a glass cloth, and drying it in an oven at 120 °C to 180 °C for a curing period of the prepreg being between 60 and 180 seconds to make it into a semi-cured state (B-stage).

[0041] A laminated board for a printed wiring board according to the present invention can be manufactured, for example, by stacking a prescribed number of prepreg sheets as mentioned above and hot-pressing them at 140°C to 200 °C and at 0.98 MPa to 4.9 MPa (mega Pascal) by laminate molding.

[0042] In this process, a metal foil is placed on one or both sides of a prescribed number of the prepreg sheets stacked for printed wiring board and laminate-molded to yield a metal foil-clad laminated board, which is fabricated into a printed wiring board. As the metal foil in this product, there may be used a copper foil, a silver foil, an aluminum foil, a stainless steel foil or the like.

[0043] In addition, a circuit is formed (formation of pattern) on an outer layer of the laminated board for printed wiring board manufactured as above to yield a printed wiring board according to the present invention. Specifically, for example, formation of the circuit can be achieved by application of a subtractive method to the metal foil on the outer layer of the metal clad laminated board to complete the printed wiring board.

[0044] Furthermore, a laminated printed wiring board can be produced through laminate-molding, using at least any one of a prepreg for printed wiring board, a laminated board for printed wiring board and a printed wiring board. Specifically, for example, a printed wiring board, which is completed with a circuit formation as above is used as a substrate board for an inner layer and a desired number of the prepreg sheets for printed wiring board are placed on one or both sides of this substrate board for the inner layer, as well as a metal foil is placed on the outside such that the metal foil side of the metal foil faces outwards, followed by hot-pressing them by laminate molding to yield a multilayered laminated board. In this process, the molding temperature is preferably set at a range between 150 °C and 180°C.

[0045] In addition, after the metal foil is coated by a bar coater with the epoxy resin composition in the form of varnish, the composition is dried at 160°C for approximately 10 minutes to yield a resin-coated metal foil.

EXAMPLE

[0046] The present invention is specifically described using following examples.

[0047] At first, epoxy resins, curing agents, silica fillers, curing accelerator, additive, and organic solvents used are given in this order.

The epoxy resins used are as follows:

Epoxy 1: Dainippon Ink and Chemical Industries, Ltd., Epiclon153-60M, epoxy equivalent = 400g/eq and bromine content = 47% (brominated epoxy resin)

Epoxy 2: Shell Chemicals Co., EPON1124-A-80, epoxy equivalent = 435 g/eq and bromine content = 19.5% (epoxy resin obtained by reacting a bifunctional phenol derivative with a bisphenol A type epoxy resin.)

Epoxy 3: Dainippon Ink and Chemical Industries, Ltd., Epiclon1120-80M, epoxy equivalent = 485 g/eq and bromine content = 20% (epoxy resin obtained by reacting a dihydric phenol with a bishphenol A type epoxy resin.)

Epoxy 4: Nippon Kayaku Co., BREN-S, epoxy equivalent = 285 g/eq and bromine content = 35.5% (brominated epoxy resin.)

Epoxy 5: Toto Kasei Co., YDCN-703, epoxy equivalent = 210 g/eq (novolac type epoxy resin)

Epoxy 6: Dainippon Ink and Chemical Industries, Ltd., HP-7200H, epoxy equivalent = 280 g/eq (dicyclopentadiene type epoxy resin)

Epoxy 7: Dainippon Ink and Chemical Industries, Ltd., Epiclon 850, epoxy equivalent = 190 g/eq (bisphenol A type epoxy resin)

Epoxy 8: Dainippon Ink and Chemical Industries, Ltd., Epiclon N660, epoxy equivalent = 210 g/eq (cresol novolac type epoxy resin)

Curing agents used are as follows:

Curing agent 1: Japan Epoxy Resin Co., YLH129B70, bishpenol A type novolac, equivalent of hydroxyl group = 118 g/eq

Curing agent 2: Dainippon Ink and Chemical Industries, Ltd., TD-2093, phenol novolac, equivalent of hydroxyl group = 105 g/eq

Curing agent 3: Dainippon Ink and Chemical Industries, Ltd., VH-4170, bisphenol A type novolac, equivalent of hydroxyl group = 118 g/eq

Curing agent 4: Dainippon Ink and Chemical Industries, Ltd, TD-2090, phenol novolac, equivalent of hydroxyl group = 105 g/eq

Curing agent 5: Gun Ei Chemical Industry Co., RESITOP PSM-4324, phenol novolac, equivalent of hydroxyl group = 105 g/eq

Curing agent 6: Dainippon Ink and Chemical Industries, Ltd., LA-7052, denatured phenol novolac, equivalent of hydroxyl group = 120 g/eq.

Silica fillers used are as follows:

Silica filler 1: Denki Kagaku Kogyo Co., FS-2DC, treated with heat, average particle diameter = 2.0 μm, specific surface area = 11.4 m^2/g, geometry crushed, and electric conductivity = 5.1 μs/cm.

Silica filler 2: Tatsumori Co., Krystallite 5X, not treated with heat, average particle diameter = 1.5μm, specific surface area = 16.5 m^2/g, geometry crushed, and electric conductivity = 2 μs/cm

Silica filler 3: Tatsumori Co., Fuserex AS-1, treated with heat, average particle diameter = 3.0 μm, relative surface area = 16.2 m^2/g, geometry crushed, and electric conductivity = 1 μs/cm

Silica filler 4: Tatsumori Co., Krystallite WX, treated with heat, average particle diameter = 1.2 μm, specific surface area = 15.3 m^2/g, geometry crushed, and electric conductivity = 7 μs/cm

Silica filler 5: Denki Kagaku Kogyou Co., FS-30, treated with heat, average particle diameter = 6.1 μm, specific surface area = 4.5 m^2/g, geometry crushed, and electric conductivity = 2.8 μs/cm

Silica filler 6: ADAMATECHS Co., SO-C2, treated with heat, average particle diameter = 0.5 μm, relative surface area = 8 m^2/g, geometry spherical and electric conductivity = 7.8 μs/cm

[0048] In addition, the specific surface area is measured with a BET method; whereas the average particle diameter is determined by a laser diffraction method to be referred to a d50 value. The electric conductivity is also determined with a conductivity cell measuring the electric conductivity of an extracted water of 10 g of the sample added to 100 ml of purified water and then pulsated for 30 minutes.
Curing accelerator used is as follows:

Accelerator 1: Shikoku Chemicals Corp., 2-ethyl-4-methyoimidazole. Additive used is as follows:

Additive 1: Daihachi Chemical Industry Co., PX-200 (fire retardant): Organic solvents used are as follows:

Organic solvent 1: methyl ethyl ketone

Organic solvent 2: methoxypropanol

[0049] EXAMPLES 1 TO 10 AND COMPARATIVE EXAMPLES 1 TO 3
[0050] The materials given in Table 1 were formulated in a desired amount (unit, pbw) and stirred for approximately 90 minutes, and then a silica filler was uniformly dispersed in a varnish with a NanoMill grinding mill to yield an epoxy resin composition (varnish) for printed wiring board in Examples 1 to 10 and in Comparative Examples 1 to 3.
[0051] In addition, a bromine content in the epoxy resin composition was calculated by the following method:
[0052] A sum of the bromine content in each epoxy resin multiplied with the amount of the solid content formulated is divided by a sum of the solid content formulated with both each epoxy resin and a curing agent, then multiplied by 100.
(Manufacturing method of prepreg for printed wiring board)
[0053] A glass cloth 0.2 mm thick (Nitto Boseki Co., "WEA7628") was impregnated with the varnish of the resin composition for printed wiring board obtained in Examples 1 to 10 and in Comparative Examples 1 to 3 and dried in a drying oven (120 °C to 180 °C) for a curing period between 60 seconds and 180 seconds to adjust the amount of the resin impregnated being either 40% or 50% by weight, yielding a prepreg for printed wiring board in a half-cured state (B-stage).

MANUFACTURING METHOD OF LAMINATED BOARD FOR PRINTED WIRING BOARD

[0054] Four or five sheets of the products of prepreg for printed wiring board containing 40% by weight impregnated resin, obtained as mentioned above, were chosen, on both sides of which a copper foil was placed, followed by hot-pressing them at 140 °C to 180 °C under 0.98 MPa to 3.9 MPa with a pressing machine by laminate molding to yield a copper-clad laminated board with board thickness of either 0.8 mm or 1.0 mm. The heating period during the laminate molding was set such that the temperature of the prepreg for printed wiring board as a whole reached 160 °C or higher for at least 60 minutes or longer. Furthermore, as the copper foil, a "GT" foil from Furukawa Circuit Foil Co. (thickness, 18 μm) was used.
[0055] Physical properties listed below of the prepreg for printed wiring board and the laminated board for printed wiring board thus obtained were evaluated. The results are shown in Table 2.

APPEARANCE OF PREPREG

[0056] Appearance of the products of prepreg for printed wiring board containing 50% by weight impregnated resin, obtained by the manufacturing method as above was visually inspected.

GLASS TRANSITION TEMPERATURE

[0057] After a copper foil of the laminated board for printed wiring board obtained in the above was removed by etching, the specimen was used to measure the glass transition temperature by a differential scanning calorimetric method according to IPC-TM-650 section 2.4.25.

FIRE RETARDANCY

[0058] After a surface copper foil of the copper clad laminated board with a board thickness of 0.8 mm was removed by etching, the specimen was cut into 125 mm long and 13 mm wide and burned by a UL vertical burning test method (UL94).

MEASUREMENT OF CURING TIME

[0059] The prepreg prepared as above was rubbed to loosen to a powder, which was filtered through a 60 mesh filter in order to remove foreign materials such as glass fiber, and then its curing period of time was measured according to JIS-C6521, section 5.7.

MEASUREMENT OF PERCENT OF WATER ABSORPTION

[0060] After the surface copper foil of the copper clad laminated board with board thickness of 1.0 mm was removed by etching, the board was cut into a 50 mm square and conditioned under a pre-conditioning time (PCT) of 3 hours and 3 atmospheric pressure to determine the percent of water adsorption, which was then calculated by the equation below:
[0061]

$$\text{Percent of water absorption} = ((\text{weight after processing} - \text{initial weight}) / \text{initial weight}) \times 100\ (\%)$$

PEAL STRENGTH OF COPPER FOIL

[0062] Peal strength of the copper foil was measured with use of the copper-clad laminated board with board thickness of 1.0 mm according to JIS-C6481.

RESULTS OF EVALUATION

[0063] It is clearly illustrated in Table 2 that the appearance of the prepreg in Examples 1 to 10, in which a silica filler contained has at least two or more planes in the geometry, an average particle diameter between 0.3□ and 10□, and a specific surface area between 8 m^2/g and 30 m^2/g (preferably between 10 m^2/g and 20 m^2/g), is better than that in Comparative Example 1 containing no silica filler and in Comparative Examples 2 and 3 containing silica filler outside the scope specified above.

Table 1

Composition		Example 1	Example 2	Example 3	Example 4	Example 5
Epoxy resin	Epoxy 1		41.7	16.7	66.7	
	Epoxy 2			112.5		112.5
	Epoxy 3	112.5				
	Epoxy 4		25.0			
	Epoxy 5	10.0			60.0	10.0
	Epoxy 6		50.0			
	Epoxy 7					
	Epoxy 8					
Curing agent	Curing agent 1			41.1		
	Curing agent 2				40.5	
	Curing agent 3	27.6				
	Curing agent 4		34.6			

	Curing agent 5					26.7
	Curing agent 6					
Additive	Additive 1					
Curing accelerator	Accelerator 1	0.128	0.067	0.128	0.070	0.060
Silica filler	Silica filler 1				70.3	
	Silica filler 2	25.5				
	Silica filler 3		26.9			31.7
	Silica filler 4			19.3		
	Silica filler 5					
	Silica filler 6					
Organic solvent	Organic solvent 1	17.0	18.0	—	25.0	22.7
	Organic solvent 2	26.0	38.2	28.0	25.0	22.7
Bromine content (%) (per whole amount of resin solid content		14.1%	15.5%	17.4%	13.7%	13.9%

Composition		Example 6	Example 7	Example 8	Example 9	Example 10
Epoxy resin	Epoxy 1	41.7			41.7	66.7
	Epoxy 2					
	Epoxy 3					
	Epoxy 4	25.0			25.0	
	Epoxy 5					60.0
	Epoxy 6	50.0			50.0	
	Epoxy 7		100.0			
	Epoxy 8			100.0		
Curing agent	Curing agent 1					

	Curing agent 2				38.8	
	Curing agent 3					
	Curing agent 4	34.6	55.3			40.5
	Curing agent 5					
	Curing agent 6			95.2		
Additive	Additive 1		40.0			
Curing accelerator	Accelerator 1	0.067	0.150	0.120	0.070	0.056
Silica filler	Silica filler 1		46.6			
	Silica filler 2			15.7	111.0	
	Silica filler 3					
	Silica filler 4	13.5				182.6
	Silica filler 5					
	Silica filler 6					
Organic solvent	Organic solvent 1	18.0	75.0	43.0	45.0	50.0
	Organic solvent 2	38.2	25.0	—	45.0	60.0
Bromine content (%) (per whole amount of solid content		15.5%	0.0%	0.0%	15.0%	13.7%

Composition		Comparative example 1	Comparative example 2	Comparative example 3
Epoxy resin	Epoxy 1			
	Epoxy 2			
	Epoxy 3	112.5	112.5	112.5
	Epoxy 4			

	Epoxy 5	10.0	10.0	10.0
	Epoxy 6			
	Epoxy 7			
	Epoxy 8			
Curing agent	Curing agent 1			
	Curing agent 2			
	Curing agent 3	27.6	27.6	27.6
	Curing agent 4			
	Curing agent 5			
	Curing agent 6			
Additive	Additive 1			
Curing accelerator	Accelerator 1	0.128	0.067	0.128
Silica filler	Silica filler 1			
	Silica filler 2			
	Silica filler 3			
	Silica filler 4			
	Silica filler 5		25.5	
	Silica filler 6			25.5
Organic solvent	Organic solvent 1	18.5	17.0	17.0
	Organic solvent 2	18.5	26.0	26.0
Bromine content (%) (per whole amount of resin solid content		14.1%	14.1%	14.1%

Note

	Name of manufacture	Trade name	Epoxy equivalent	Bromine content	Remarks
Epoxy 1	Dainippon Ink and Chemical Industries, Ltd	Epiclon 153-60M	400 g/eq	48.0%	Brominated epoxy resin
Epoxy 2	Shell Chemical Co.	EPON 1124-A-80	435 g/eq	19.5%	Epoxy resin obtained by reacting a dihydric phenol with bisphenol A type epoxy resin
Epoxy 3	Dainippon Ink and Chemical Industries, Ltd	Epiclon 1120-80M	485 g/eq	20.0%	Epoxy resin obtained by reacting a dihydric phenol with bisphenol A type epoxy resin
Epoxy 4	Nippon Kayaku Co.	BREN-S	285 g/eq	35.5%	Brominated epoxy resin
Epoxy 5	Toto Kasei Co.	YDCN-703	210 g/eq	—	Novolac type epoxy resin
Epoxy 6	Dainippon Ink and Chemical Industries, Ltd	HP-7200H	280 g/eq	—	Dicyclopentadiene type epoxy resin
Epoxy 7	Dainippon Ink and Chemical Industries, Ltd	Epiclon 850	190 g/eq	—	Bisphenol A type epoxy resin
Epoxy 8	Dainippon Ink and Chemical Industries, Ltd	Epiclon N660	210 g/eq	—	Cresol novolac type epoxy resin
	Name of manufacture	Trade name	Equivalent of hydroxyl group		Remark
Curing agent 1	Japan Epoxy Resin Co.	YLH129B70	118 g/eq		Bisphenol A type novolac
Curing agent 2	Dainippon Ink and Chemical Industries, Ltd.	TD-2093	105 g/eq		Phenol novolac
Curing agent 3	Dainippon Ink and Chemical Industries, Ltd.	VH-4170	118 g/eq		Bisphenol A novolac
Curing agent 4	Dainippon Ink and Chemical Indus-	TD-2090	105 g/eq		Phenol novolac

	tries, Ltd.				
Curing agent 5	Gun Ei Chemical Industry Co.	Resitop PSM-4324	105 g/eq		Phenol novolac
Curing agent 6	Dainippon Ink and Chemical Industries, Ltd.	LA-7052	120 g/eq		Modified phenol novolac
	Name of manufacture	Trade name			
Additive 1	Daihachi Chemical Industry Co.	PX-200 (fire retardant)			
	Name of manufacture	Trade name			
Accelerator 1	Shikoku Chemical Co.	2-Ethyl-4-methyl-imidazole			

	Name of manufacture	Trade name	Heat treatment	Average particle diameter	Specific surface area	Geometry	Electric conductivity
Silica filler 1	Denki Kagaku Kogyo Co.	FS-2DC	yes	2.0 µm	11.4 m^2/g	Crushed	5.1 µs/cm
Silica filler 2	Tatsumori Co.	Krystalite 5X	no	1.5 µm	16.5 m^2/g	Crushed	2.0 µs/cm
Silica filler 3	Tatsumori Co	Fuserex AS-1	yes	3.0 µm	16.2 m^2/g	Crushed	1.0 µs/cm
Silica filler 4	Tatsumori Co	Fusesrex WX	yes	1.2 µm	15.3 m^2/g	Crushed	7.0 µs/cm
Silica filler 5	Denki Kagaku Kogyo Co.	FS-30	yes	6.1 µm	4.5 m^2/g	Crushed	2.8 µs/cm
Silica filler 6	ADNATECHS Co.	SO-C2	yes	0.5 µm	8.0 m^2/g	spherical	7.8 µs/cm

Organic solvent 1	Methyl ethyl ketone
Organic solvent 2	Methoxypropanol

Table 2

		Example 1	Example 2	Example 3	Example 4	Example 5
Silica filler	Amount added	20 parts	20 parts	15 parts	50 parts	25 parts
	Average particle diameter (□)	1.5	3.0	1.2	2.0	3.0
	Specific surface area (m^2/g)	16.5	16.2	15.3	11.4	16.2
	Geometry	crushed	crushed	crushed	crushed	crushed
	Heat treatment	no	yes	yes	yes	yes
	Electric conductivity (□/cm)	2.0	1.0	7.0	5.1	1.0
Evaluation results	Appearance of prepreg	good	good	good	good	good
	Fire retardancy	V-0	V-0	V-0	V-0	V-0
	Tg (DSC)	137 °C	165 °C	135 °C	175 °C	140 °C
	Percent of water absorption (%)	0.47	0.31	0.51	0.30	0.45
	Peal strength of copper foil (KN/m^2)	1.2	1.1	1.2	1.1	1.2

		Example 6	Example 7	Example 8	Example 9	Example 10
Silica filler	Amount added	10 parts	30 parts	10 parts	80 parts	130 parts
	Average particle diameter (□)	1.2	2.0	3.0	1.5	1.2
	Specific surface area (m^2/g)	15.3	11.4	16.2	16.5	15.3
	Geometry	crushed	crushed	crushed	crushed	crushed
	Heat treatment	yes	yes	yes	no	yes
	Electric conductivity (□/cm)	7.0	5.1	1.0	2.0	7.0
Evaluation results	Appearance of prepreg	good	good	good	good	good
	Fire retardancy	V-0	V-0	V-0	V-0	V-0
	Tg (DSC)	165 °C	113 °C	160 °C	165 °C	175 °C
	Percent of water absorption (%)	0.36	0.61	0.40	0.25	0.20

	Peal strength of copper foil (KN/m^2)	1.1	1.1	1.1	1.1	0.8

		Comparative Example 1	Comparative Example 2	Comparative Example 3
Silica filler	Amount added	none	20 parts	20 parts
	Average particle diameter (□)	—	6.1	0.5
	Specific surface area (m^2/g)	—	4.5	8.0
	Geometry	—	crushed	Spherical
	Heat treatment	—	yes	yes
	Electric conductivity (□/cm)	—	2.8	7.8
Evaluation results	Appearance of prepreg	Sagging of resin	Sagging of resin	Sagging of resin
	Fire retardancy	V-0	V-0	V-0
	Tg (DSC)	137 °C	137 °C	137 °C
	Percent of water absorption (%)	0.60	0.47	0.47
	Peal strength of copper foil (KN/m^2)	1.2	1.2	1.2

INDUSTRIAL APPLICABILITY

[0064] The epoxy resin composition for printed wiring board according to the present invention, which uses, in a resin composition comprised of an epoxy resin, a phenol novolac resin and a curing accelerator, a silica filler having a geometry possessing at least two planes, an average particle diameter between 0.3 μm and 10 μm and a specific surface area between 8 m^2/g and 30 m^2/g, is capable of increasing apparent resin viscosity to prevent resin sagging in a drying oven and capable of penetrating into a reinforcing material without being impaired because viscosity of the resin itself is locally minimally increased, leading to improvement of the appearance of the prepreg.

[0065] In the epoxy resin composition for printed wiring board according to the invention, through addition of a silica filler possessing at least two planes, an average particle diameter between 0.3 μm and 10 μm, and a specific surface area between 10 m^2/g and 20 m^2/g to the resin in an amount between 3% and 80% by weight per the solid content of the resin, there can be attained effects of increasing apparent resin viscosity to prevent resin sagging in a drying oven and of improving appearance of the prepreg without impairing penetration of the resin into the reinforcing material, because viscosity of the resin itself is not locally increased.

[0066] The epoxy resin composition for printed wiring board according to this invention gives excellent reliability in a long term electric insulation, by using, as the silica filler, ones having an electric conductivity of 15 μs/cm or less.

[0067] The epoxy resin composition for printed wiring board according to the invention is capable of eliminating crystalline substances as hazardous materials, by using, as the silica filler, vitrified ones through melting the silica at a temperature of 1800 °C or higher.

Claims

1. An epoxy resin composition for a printed wiring board, comprising an epoxy resin, a phenol novolac resin, a curing accelerator and a silica filler, **characterized in that**, as the silica filler, is used a silica filler which has a shape having at least two planes, and has an average particle diameter between 0.3 μm and 10 μm and a relative surface area measured with BET between 8 m^2/g and 30 m^2/g.

2. An epoxy resin composition for a printed wiring board as described in claim 1, **characterized in that**, as said silica filler defined in claim 1, is used a silica filler having at least two planes in the shape, an average particle diameter between 0.3 μm and 10 μm and a relative surface area between 10 m^2/g and 20 m^2/g.

3. An epoxy resin composition for a printed wiring board as described in claims 1 or 2, **characterized in that** said silica filler defined in claim 1 or 2 is added in an amount of from 3% to 80% by weight per the solid content of the resin.

4. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 3, **characterized in that**, as said silica filler defined in claim 1, is used a silica filler having an electric conductivity of 15 μs or less, whereby the electric conductivity is determinend with a conductivity cell measuring the electric conductivity of an extracted water of 10 g of a sample added to 100 ml of purified water and then pulsated for 30 minutes.

5. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 4, **characterized in that**, as said silica filler defined in claim 1 or 2, is used a silica filler which has been vitrified through melting at a temperature of 1800 °C or higher.

6. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 5, **characterized in that**, as said epoxy resin, is used an epoxy resin having a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler and containing an epoxy resin obtained by reacting a dihydric phenol with a bisphenol A type epoxy resin in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

7. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 5, **characterized in that**, as said epoxy resin, is used an epoxy resin having a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler and containing an epoxy resin possessing a dicyclopentadienyl structure in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

8. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 5, **characterized in that**, as said epoxy resin, is used an epoxy resin having a bromine content of between 5% and 20% by weight per the solid content of the resin without silica filler and containing of a novolac type epoxy resin in an amount of between 40% and 100% by weight based on the whole amount of the epoxy resin solid content.

9. An epoxy resin composition for a printed wiring board as described in any one of claims 1 to 5, **characterized in that**, as said epoxy resin composition, is used a bromine-free epoxy resin composition.

10. A process [prepreg] for producing a printed wiring board, which comprises using a prepreg in producing a printed wiring board; said [**characterized in that** the] prepreg is obtained by impregnating a reinforcing material with an epoxy resin composition for a printed wiring board [as described in any one of claims 1 to 9] and drying said composition to B-stagesaid epoxy resin composition comprising an epoxy resin, a phenol novolac resin, a curing accelerator, and a silica filler which has a shape having at least two planes and has an average particle diameter between 0.3 μm and 10 μm and a relative surface area measured by BET between 8 m^2/g and 30 m^2/g.

11. A process [laminated board] for producing a printed wiring board, which comprises using a laminate board in producing a printed wiring board; said laminate [**characterized in that** the] board is obtained by gluing a prepreg as described in claim 10 to a surface of a metal foil and hot pressing them.

12. A printed wiring board, which [**characterized in that** the board] is obtained by using a laminated board for a printed wiring board [as described in claim 11] ; said laminate board is obtained by gluing a prepreg as described in claim 10 to a surface of a metal foil and hot pressing them.

13. A process for producing a printed wiring board as described in claim 1, which comprises the following steps:

(1) preparing a prepreg for a printed wiring board, by preparing a varnish of said epoxy resin composition with

an organic solvent, impregnating the varnish into a glass cloth, and drying it in an oven to make it into a semi-cured state (B-stage);
(2) manufacturing s laminated board for a printed wiring board, by stacking a prescribed number of sheets of said prepreg and hot-pressing them through laminate molding, and placing a metal foil on one or both sides of the prescribed number of the prepreg sheets stacked for printed wiring board and laminate-molded to yield a metal foil-clad laminated board; and
(3) forming a circuit on an outer layer of the laminated board for printed wiring board to yield a printed wiring board.

14. A process for producing a printed wiring board, which comprises the following steps:

(1) preparing a prepreg for a printed wiring board, by preparing a varnish of said epoxy resin composition with an organic solvent, impregnating the varnish into a glass cloth, and drying it in an oven to make it into a semi-cured state (B-stage); wherein said epoxy resin composition comprises an epoxy resin, a phenol novolac resin, a curing accelerator, and a silica filler having a shape having at least two planes and having an average particle diameter between 0.3 μm and 10 μm and a relative surface area measured by BET between 8 m^2/g and 30 m^2/g;
(2) manufacturing s laminated board for a printed wiring board, by stacking a prescribed number of sheets of said prepreg and hot-pressing them through laminate molding, and placing a metal foil on one or both sides of the prescribed number of the prepreg sheets stacked for printed wiring board and laminate-molded to yield a metal foil-clad laminated board; and
(3) forming a circuit on an outer layer of the laminated board for printed wiring board to yield a printed wiring board.

Patentansprüche

1. Epoxidharz-Zusammensetzung für eine Leiterplatte, umfassend ein Epoxidharz, ein Phenolnovolakharz, einen Härtungsbeschleuniger und einen Quarzfüllstoff, **dadurch gekennzeichnet, dass** als Quarzfüllstoff ein Quarzfüllstoff verwendet wird, der eine Form mit mindestens zwei Ebenen aufweist, dessen mittlerer Teilchendurchmesser zwischen 0,3 μm und 10 μm liegt und dessen mit BET gemessene relative Fläche zwischen 8 m^2/g und 30 m^2/g beträgt.

2. Epoxidharz-Zusammensetzung für eine Leiterplatte nach Anspruch 1, **dadurch gekennzeichnet, dass** als in Anspruch 1 definierter Quarzfüllstoff ein Quarzfüllstoff verwendet wird, der eine Form mit mindestens zwei Ebenen aufweist, dessen mittlerer Teilchendurchmesser zwischen 0,3 μm und 10 μm liegt und dessen relative Fläche zwischen 10 m^2/g und 20 m^2/g beträgt.

3. Epoxidharz-Zusammensetzung für eine Leiterplatte nach Anspruch 1 oder 2, **dadurch gekennzeichnet, dass** der in Anspruch 1 oder 2 definierte Quarzfüllstoff in einer Menge von zwischen 3 und 80 Gew.-% pro Feststoffgehalt des Harzes hinzugefügt wird.

4. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 3, **dadurch gekennzeichnet, dass** als in Anspruch 1 definierter Quarzfüllstoff ein Quarzfüllstoff verwendet wird, der eine elektrische Leitfähigkeit von 15 μs oder weniger aufweist, wobei die elektrische Leitfähigkeit mit einer Leitfähigkeitsmesszelle bestimmt wird, die die elektrische Leitfähigkeit von aus 10 g einer Probe abgezogenem Wasser misst, welches 100 ml gereinigtem Wasser zugesetzt und dann 30 Minuten lang gepulst wird.

5. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 4, **dadurch gekennzeichnet, dass** als in Anspruch 1 oder 2 definierter Quarzfüllstoff ein Quarzfüllstoff verwendet wird, der bei einer Temperatur von 1800 °C oder mehr zu Glas geschmolzen wird.

6. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 5, **dadurch gekennzeichnet, dass** als Epoxidharz ein Epoxidharz mit einem Bromgehalt von zwischen 5 und 20 Gew.-% pro Feststoffgehalt des Harzes ohne Quarzfüllstoff verwendet wird, das ein Epoxidharz enthält, das durch Reaktion eines dihydrischen Phenols mit einem Epoxidharz des Typs Bisphenol A in einer Menge von zwischen 40 und 100 Gew.-% bezogen auf die Gesamtmenge des Feststoffgehalts des Epoxidharzes erzielt wird.

7. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 5, **dadurch gekennzeichnet, dass** als das Epoxidharz ein Epoxidharz mit einem Bromgehalt von zwischen 5 und 20 Gew.-% pro Feststoffgehalt des Harzes ohne Quarzfüllstoff verwendet wird, das ein Epoxidharz enthält, das eine Dicyclopentadienyl-Struktur in einer Menge von zwischen 40 und 100 Gew.-% bezogen auf die Gesamtmenge des Feststoffgehalts des Epoxidharzes besitzt.

8. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 5, **dadurch gekennzeichnet, dass** als das Epoxidharz ein Epoxidharz mit einem Bromgehalt von zwischen 5 und 20 Gew.-% pro Feststoffgehalt des Harzes ohne Quarzfüllstoff verwendet wird, das ein Epoxidharz des Typs Novolak in einer Menge von zwischen 40 und 100 Gew.-% bezogen auf die Gesamtmenge des Feststoffgehalts des Epoxidharzes enthält.

9. Epoxidharz-Zusammensetzung für eine Leiterplatte nach einem der Ansprüche 1 bis 5, **dadurch gekennzeichnet, dass** as Epoxidharz-Zusammensetzung eine Brom freie Epoxidharz-Zusammensetzung verwendet wird.

10. Verfahren [Prepreg] zur Herstellung einer Leiterplatte, das darin besteht, ein Prepreg bei der Herstellung einer Leiterplatte zu verwenden; wobei [**dadurch gekennzeichnet, dass**] das Prepreg **dadurch** erzielt wird, dass ein Verstärkungsmaterial mit einer Epoxidharz-Zusammensetzung für eine Leiterplatte [nach einem der Ansprüche 1 bis 9] imprägniert wird, und dass diese Zusammensetzung getrocknet wird, um eine B-Stufe zu erzielen; diese Epoxidharz-Zusammensetzung umfasst ein Epoxidharz, ein Phenolnovolakharz, einen Härtungsbeschleuniger, und einen Quarzfüllstoff, der eine Form mit mindestens zwei Ebenen und einen mittleren Teilchendurchmesser von zwischen 0,3 μm und 10 μm aufweist und dessen mit BET gemessene relative Fläche zwischen 8 m^2/g und 30 m^2/g beträgt.

11. Verfahren [Laminatplatte] zur Herstellung einer Leiterplatte, das darin besteht, eine Laminatplatte bei der Herstellung einer Leiterplatte zu verwenden; wobei [**dadurch gekennzeichnet, dass**] die Laminatplatte **dadurch** erzielt wird, dass ein Prepreg nach Anspruch 10 auf eine Fläche einer Metallfolie aufgeklebt wird und diese beiden heiß verpresst werden.

12. Leiterplatte, die [**dadurch gekennzeichnet** wird, dass sie] erhalten wird durch Verwendung einer Laminatplatte für eine Leiterplatte [nach Anspruch 11], wobei die Laminatplatte **dadurch** erzielt wird, dass ein Prepreg nach Anspruch 10 auf eine Fläche einer Metallfolie aufgeklebt wird und diese beiden heiß verpresst werden.

13. Verfahren zur Herstellung einer Leiterplatte nach Anspruch 11, umfassend folgende Schritte:

(1) Zubereiten eines Prepreg für eine Leiterplatte durch Zubereitung eines Lackes aus der besagten Epoxidharz-Zusammensetzung mit einem organischen Lösungsmittel, Imprägnieren eines Glasgewebes mit dem Lack und Trocknen desselben in einem Ofen bis zu einem halbgehärteten Zustand (B-Stufe);
(2) Herstellen einer Laminatplatte für eine Leiterplatte durch Aufeinanderstapeln einer vorgeschriebenen Anzahl an Lagen dieses Prepreg und durch Heißverpressen derselben durch gleichzeitiges Laminieren, Formen und Härten und Aufbringen einer Metallfolie auf eine oder beide Seiten der vorgeschriebenen Anzahl an für die Leiterplatte gestapelten Prepreglagen, die zur Erzielung einer mit einer Metallfolie überzogenen Laminatplatte zu einem Laminat formgepresst werden; und
(3) Bilden einer Schaltung auf einer Außenlage der Laminatplatte für die Leiterplatte zur Erzielung einer Leiterplatte.

14. Verfahren zur Herstellung einer Leiterplatte, umfassend folgende Schritte:

(1) Zubereiten eines Prepreg für eine Leiterplatte durch Zubereitung eines Lackes aus der besagten Epoxidharz-Zusammensetzung mit einem organischen Lösungsmittel, Imprägnieren eines Glasgewebes mit dem Lack und Trocknen desselben in einem Ofen bis zu einem halbgehärteten Zustand (B-Stufe), wobei die besagte Epoxidharz-Zusammensetzung ein Epoxidharz, ein Phenolnovolak-Harz, einen Härtungsbeschleuniger, und einen Quarzfüllstoff umfasst, der eine Form mit mindestens zwei Ebenen und einen mittleren Teilchendurchmesser von zwischen 0,3 μm und 10 μm aufweist und dessen mit BET gemessene relative Fläche zwischen 8 m^2/g und 30 m^2/g beträgt;
(2) Herstellen einer Laminatplatte für eine Leiterplatte durch Aufeinanderstapeln einer vorgeschriebenen Anzahl an Lagen dieses Prepreg und durch Heißverpressen derselben durch Formpressen eines Laminates und Aufbringen einer Metallfolie auf eine oder beide Seiten der vorgeschriebenen Anzahl an für die Leiterplatte gestapelten Prepreglagen, die zur Erzielung einer mit einer Metallfolie überzogenen Laminatplatte zu einem Laminat formgepresst werden; und
(3) Bilden einer Schaltung auf einer Außenlage der Laminatplatte für die Leiterplatte zur Erzielung einer Leiterplatte.

Revendications

1. Composition de résine époxy pour carte de circuit imprimé, du type comportant une résine époxy, une résine novolaque phénol, un accélérateur de prise et une charge de silice, **caractérisée en ce que**, comme charge de silice, on utilise une charge de silice ayant une forme avec au moins deux plans et un diamètre moyen de particules compris entre 0,3 μm et 10 μm ainsi qu'une aire relative mesurée BET comprise entre 8 m^2/g et 30 m^2/g.

2. Composition de résine époxy pour carte de circuit imprimé selon la revendication 1, **caractérisée en ce que** l'on utilise, comme charge de silice selon la revendication 1, une charge de silice ayant une forme avec au moins deux plans et un diamètre moyen de particules compris entre 0,3 μm et 10 μm ainsi qu'une aire relative mesurée BET comprise entre 10 m^2/g et 20 m^2/g.

3. Composition de résine époxy pour carte de circuit imprimé selon la revendication 1 ou 2, **caractérisée en ce que** la charge de silice selon la revendication 1 ou 2 est rajoutée dans une quantité de 3 à 80 % en poids par teneur en particules solides de la résine.

4. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 3, **caractérisée en ce que** l'on utilise, comme charge de silice selon la revendication 1, une charge de silice dont la conductivité électrique est de 15 μs ou moins, la conductivité électrique étant déterminée à l'aide d'une cellule de conductivité qui mesure la conductivité électrique de l'eau extraite d'un échantillon de 10 g ajoutée à 100 ml d'eau purifiée puis pulsée pendant 30 minutes.

5. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 4, **caractérisée en ce que** l'on utilise, comme charge de silice selon la revendication 1 ou 2, une charge de silice qui a été vitrifiée en la faisant fondre à une température de 1800 °C ou plus.

6. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 5, **caractérisée en ce que** l'on utilise, comme résine époxy, une résine époxy ayant une teneur en brome comprise entre 5 et 20 % en poids par teneur en particules solides de la résine sans charge de silice et contenant une résine époxy obtenue en faisant réagir un phénol dihydrique avec une résine époxy du type bisphénol A dans une quantité comprise entre 40 et 100 % en poids, rapportée à la teneur totale en particules solides de la résine époxy.

7. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 5, **caractérisée en ce que** l'on utilise, comme résine époxy, une résine époxy ayant une teneur en brome comprise entre 5 et 20 % en poids par teneur en particules solides de la résine sans charge de silice et contenant une résine époxy possédant une structure dicyclopentadiényle dans une quantité comprise entre 40 et 100 % en poids, rapportée à la teneur totale en particules solides de la résine époxy.

8. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 5, **caractérisée en ce que** l'on utilise, comme résine époxy, une résine époxy ayant une teneur en brome comprise entre 5 et 20 % en poids par teneur en particules solides de la résine sans charge de silice et contenant une résine époxy du type novolaque dans une quantité comprise entre 40 et 100 % en poids, rapportée à la teneur totale en particules solides de la résine époxy.

9. Composition de résine époxy pour carte de circuit imprimé selon l'une quelconque des revendications 1 à 5, **caractérisée en ce que** l'on utilise, comme composition de résine époxy, une composition de résine époxy dépourvue de brome.

10. Procédé [préimprégné] de réalisation d'une carte de circuit imprimé qui consiste à utiliser un préimprégné dans la réalisation d'une carte de circuit imprimé; ce préimprégné étant [**caractérisé en ce qu'**il est] obtenu en imprégnant un matériau de renforcement d'une composition de résine époxy pour une carte de circuit imprimé [selon l'une quelconque des revendications 1 à 9] et en séchant cette composition jusqu'à l'état B ; cette composition de résine époxy comprenant une résine époxy, une résine novolaque phénol, un accélérateur de prise et une charge de silice ayant une forme avec au moins deux plans et un diamètre moyen de particules compris entre 0,3 μm et 10 μm ainsi qu'une aire relative mesurée BET comprise entre 8 m^2/g et 30 m^2/g.

11. Procédé [carte laminée] de réalisation d'une carte de circuit imprimé, qui consiste à utiliser une carte laminée dans la réalisation d'une carte de circuit imprimé ; cette carte laminée étant [**caractérisée en ce qu'**elle est] obtenue en

collant un préimprégné selon la revendication 10 sur une surface d'une feuille de métal par pression à chaud.

12. Carte de circuit imprimé qui [**caractérisée en ce qu'**elle] est obtenue en utilisant une carte laminée pour une carte de circuit imprimé [selon la revendication 11] ; cette carte laminée étant obtenue par collage d'un préimprégné selon la revendication 10 sur une surface d'une feuille de métal par pression à chaud.

13. Procédé de réalisation d'une carte de circuit imprimé selon la revendication 11, comprenant les étapes suivantes :

(1) préparer un préimprégné pour carte de circuit imprimé en préparant, à partir de la composition de résine époxy, un vernis avec un solvant organique, en imprégnant du vernis un tissu de verre et en le séchant dans un four pour lui faire adopter un état semi-polymérisé (état B) ;
(2) réaliser une carte laminée pour une carte de circuit imprimé en empilant un nombre prescrit de feuilles de ce préimprégné et en les pressant à chaud par un procédé de laminage, de moulage et de polymérisation simultanés et en plaçant une feuille de métal sur une ou sur les deux faces du nombre prescrit de feuilles de préimprégné empilées pour former la carte de circuit imprimé et laminées, moulées et polymérisées simultanément pour obtenir une carte laminée revêtue d'une feuille de métal ; et
(3) former un circuit sur une couche extérieure de la carte laminée pour carte de circuit imprimé pour obtenir une carte de circuit imprimé.

14. Procédé de réalisation d'une carte de circuit imprimé qui comprend les étapes suivantes :

(1) préparer un préimprégné pour carte de circuit imprimé en préparant, à partir de la composition de résine époxy, un vernis avec un solvant organique, en imprégnant du vernis un tissu de verre et en le séchant dans un four pour lui faire adopter un état semi-polymérisé (état B) ; la composition de résine époxy comportant une résine époxy, une résine novolaque phénol, un accélérateur de prise et une charge de silice, **caractérisée en ce que**, comme charge de silice, on utilise une charge de silice ayant une forme avec au moins deux plans et un diamètre moyen de particules compris entre 0,3 μm et 10 μm ainsi qu'une aire relative mesurée BET comprise entre 8 m^2/g et 30 m^2/g ;
(2) réaliser une carte laminée pour carte de circuit imprimé en empilant un nombre prescrit de feuilles de ce préimprégné et en les pressant à chaud par un procédé de laminage, de moulage et de polymérisation simultanés et en plaçant une feuille de métal sur une ou sur les deux faces du nombre prescrit de feuilles de préimprégné empilées pour former la carte de circuit imprimé et laminées, moulées et polymérisées simultanément pour obtenir une carte laminée revêtue d'une feuille de métal ; et
(3) former un circuit sur une couche extérieure de la carte laminée pour carte de circuit imprimé pour obtenir une carte de circuit imprimé.

REFERENCES CITED IN THE DESCRIPTION

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