

THERMOSETTING RESIN COMPOSITION

The present application is divided out of application No. 7925679.

This invention relates to thermosetting resin compositions.

There are a variety of heat-resistant resins which can be used as insulating materials in order to meet the demands for greater capacity, smaller size, lighter weight, and higher degree of reliability of electronic devices and equipment as well as for better heat resistance characteristics, longer service life, and maintenance-free properties of electrical machinery and apparatus. Among these heat-resistant resins, imide resins of the addition polymerization type, such as bismaleimide resins and aromatic diamine-modified bismaleimide resins, are well known. Such bismaleimide resins have excellent heat resistance, but are not very satisfactory because of the difficulties in processing. The melting points of bismaleimides are high and their curing rates are very low. Moreover, their poor solubility in organic solvents necessitates the use of polar solvents, such as N-methyl-2-pyrrolidone and dimethylformamide, which are undesirable from the viewpoint of environmental pollution.

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Furthermore, an organic peroxide such as dicumyl peroxide or benzoyl peroxide is usually added to maleimide resins for the purpose of promoting their cure. When such a resin composition is used as impregnating varnish to make prepregs, the cleavage products of the organic peroxide remaining in the prepregs may lead to a reduction in quality and performance of the finally obtained laminates and, especially in the case of copper-clad laminates, to blistering or peeling-off of the copper foil.

It is well known that bismaleimides form a three-dimensional network when they are heated above their melting points or subjected to the action of free radicals. It is also known that the presence of a catalytic amount of a free-radical initiator is desirable for the purpose of achieving an effective crosslinking reaction (see, for example, J. Amer. Chem. Soc., Vol. 81, P. 1187, 1959). By nature, many free-radical initiators have a low activation energy for their decomposition and are unstable and easily decomposable even at room temperature, so that special care must be taken in their storage and handling.

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It is an object of this invention to provide an improved thermosetting resin composition.

In one aspect of the present invention there is provided a thermosetting resin composition comprising:

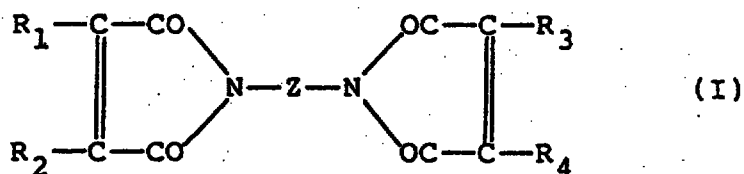
5 (a) at least one maleimide compound selected from bismaleimides and polymaleimides; and

(b) at least one alkenylphenyl compound selected from alkenylanilines and derivatives thereof, linear dimers thereof, and polymers thereof.

10 In another aspect of the present invention there is provided a thermosetting resin composition comprising:

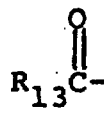
(a) at least one maleimide compound selected from

(i) bismaleimides having the formula



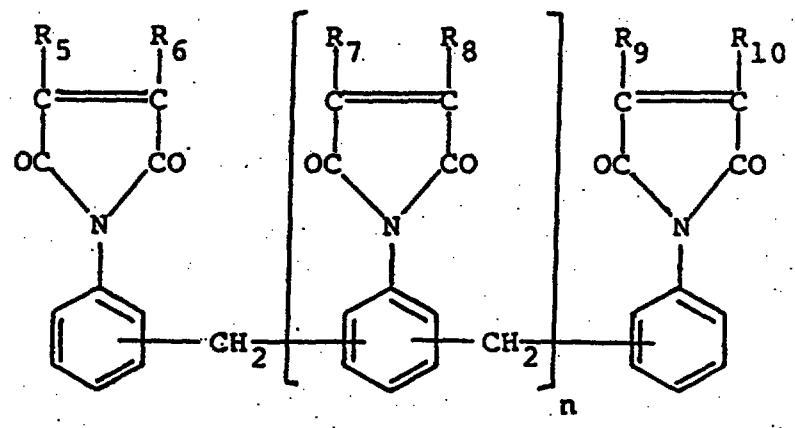
20 where R_1 , R_2 , R_3 , and R_4 are hydrogen atoms, halogen atoms; or straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals; or substituted phenyl radicals having one or more substituents selected from straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, halogen atoms, $R_{13}O-$ groups in which R_{13} is an aliphatic radical of from 1 to 5 carbon atoms, and, if there are two or more radicals represented by R_{13} , they may be identical to or different from each other,

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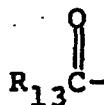
groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups; and the radicals represented by R_1 , R_2 , R_3 , and R_4 may be identical to or different from each other, and Z is a divalent organic radical of at least 2 carbon atoms; and

(ii) polymaleimides having the formula



(II)

where R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} are hydrogen atoms; halogen atoms; straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals, or substituted phenyl radicals having one or more substituents selected from halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, $R_{13}O-$ groups in which R_{13} is as previously defined,

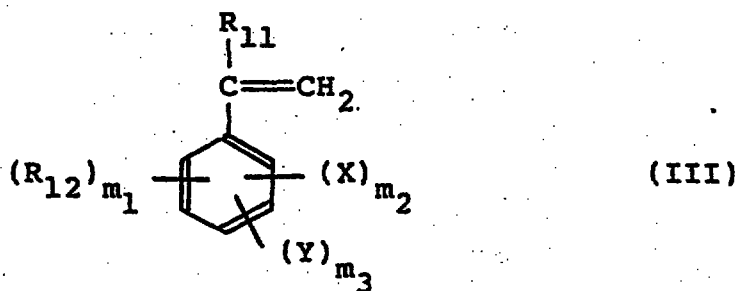


groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups, and the radicals represented by R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} may be identical to or different from each other; and n is zero or a positive number; and

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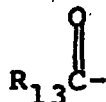
(b) at least one alkenylphenyl compound selected from

10 compounds having the formula



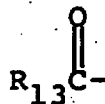
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where R_{11} and R_{12} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals, or substituted phenyl radicals having one or more substituents selected from halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, $\text{R}_{13}\text{O}-$ groups in which R_{13} is as previously defined,



25 groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups, and the radicals represented by R_{11} and R_{12} may be identical to or different from each

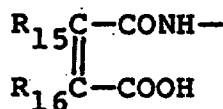
other, X is a hydrogen atom, halogen atom, carboxyl group,



group wherein R_{13} is as previously defined, $\text{R}_{13}\text{O}-$ group

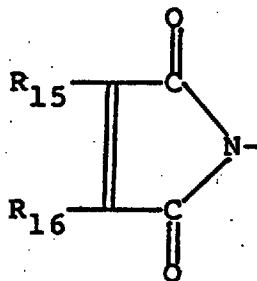
5 wherein R_{13} is as previously defined, or cyano group, Y is an amino group, glycidylamino group,

diglycidylamino group,



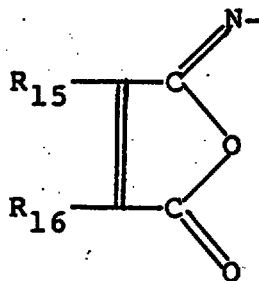
10 group in which R_{15} and R_{16} are hydrogen atoms, halogen atoms, or straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms and the radicals represented by R_{15} and R_{16} may be identical to or different from each other,

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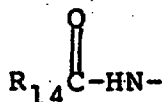


group in which R_{15} and R_{16} are as previously defined,

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25 group in which R_{15} and R_{16} are as previously defined,



group in which R_{14} is an aliphatic radical of from 1 to 20 carbon atoms and, if there are two or more radicals represented by R_{14} , they may be identical to or different from each other, alkylamino group of from 1 to 5 carbon atoms, or dialkylamino group in which each alkyl radical has from 1 to 5 carbon atoms, and m_1 , m_2 , and m_3 are positive integers and the sum of $m_1 + m_2 + m_3$ has a value of 5; linear dimers thereof; and polymers thereof.

In addition to the aforesaid components (a) and (b), this thermosetting resin composition can contain (c) at least one polymerizable unsaturated compound. The thermosetting resin compositions consisting of components (a), (b), and (c) have more excellent performance than those consisting essentially of components (a) and (b).

In a particularly preferred thermosetting resin composition of the invention in formula (I), R_1 , R_2 , R_3 , and R_4 are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 5 carbon atoms, or phenyl radicals and the radicals represented by R_1 , R_2 , R_3 , and R_4 may be identical to or different from each other, and Z is an organic radical of 2 or more carbon atoms selected from aliphatic, alicyclic, aromatic, and heterocyclic radicals and combinations thereof or a combination of an organic radical as defined above with -O-, -S-, -S-S-, -CO-, -SO-, -SO₂-, -CO₂-, -N=N-, -NH-, or -CONH-; in formula (II), R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 5

carbon atoms, or phenyl radicals and the radicals represented by R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} may be identical to or different from each other, and n has a value of from 0.1 to 10; and in formula (III), R_{11} and R_{12} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, or phenyl radicals and the radicals represented by R_{11} and R_{12} may be identical to or different from each other, X is a hydrogen or halogen atom, Y is an amino group, maleimido group, glycidylamino group, or alkylamino group of from 1 to 5 carbon atoms, and m_1 , m_2 , and m_3 are positive integers and the sum of $m_1 + m_2 + m_3$ has a value of 5.

Specific examples of the radical represented by Z in formula (I) includes two or more phenylene or cyclohexylene radicals interconnected either by a simple valence bond or by an atom



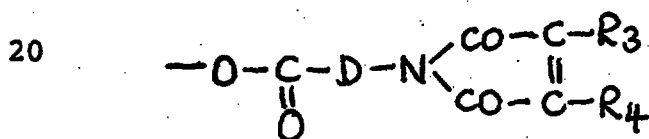
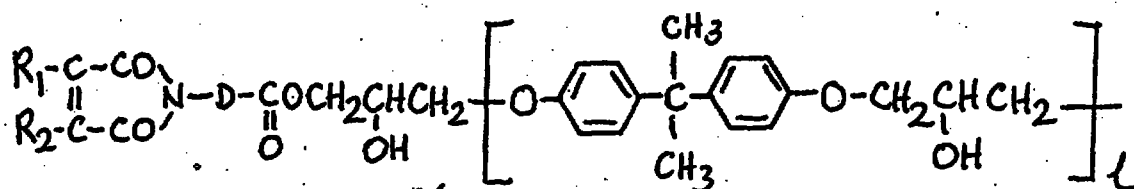
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N,N'-(thiodi-p-phenylene)bismaleimide, N,N'-(dithiodi-p-phenylene)bismaleimide, N,N'-(sulfonyldi-m-phenylene)bismaleimide, N,N'-(o,p-isopropylidenediphenylene)bismaleimide, N,N'-(isopropylidenedi-p-phenylene)bismaleimide, N,N'-(o,p-cyclohexylidenediphenylene)bismaleimide, N,N'-(cyclohexylidenedi-p-phenylene)bismaleimide, N,N'-(ethylenedi-p-phenylene)bismaleimide, N,N'-(m-xylylene)bismaleimide, N,N'-(p-xylylene)bismaleimide, N,N'-(4,4"-p-triphenylene)bismaleimide, N,N'-(p-phenylenedioxydi-p-phenylene)bismaleimide, N,N'-(methylenedi-p-phenylene)bis(2,3-dichloromaleimide), and N,N'-(oxydi-p-phenylene)bis(2-chloromaleimide). They further include maleimide-terminated phenoxy compounds having the general formula

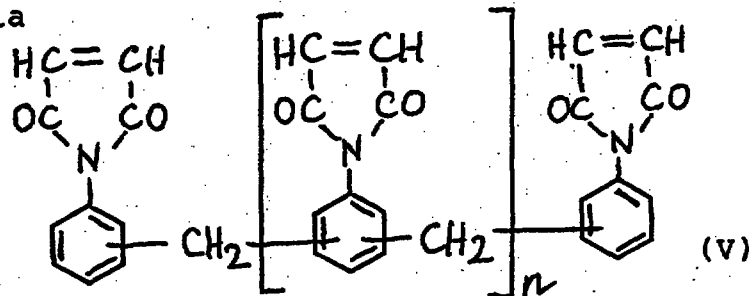


(IV)

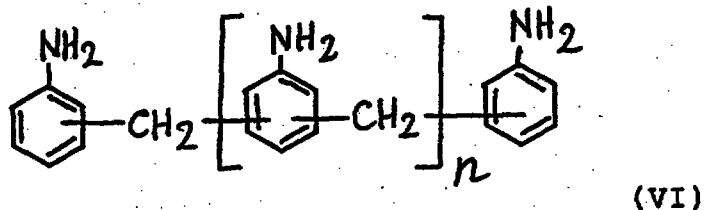
where R_1 , R_2 , R_3 , and R_4 are as previously defined for formula (I), D is a divalent organic radical selected from aliphatic, alicyclic, and aromatic radicals, and L is a number having a value from 0 to 20. Specific examples of the radical represented by D in formula (IV) include methylene, dimethylene, trimethylene, hexamethylene,

m-phenylene, and p-phenylene radicals.

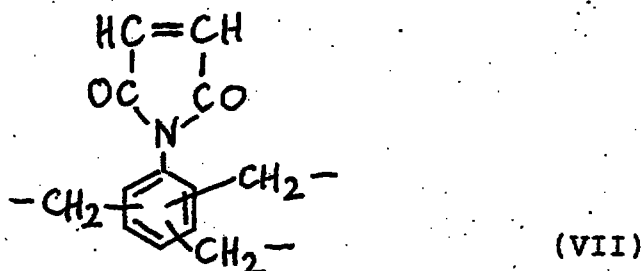
Specific examples of the compounds of formula (II) include polymethylene polyphenylene polymaleimides having the formula



where n is a number having an average value of 0 or greater. These polymaleimides can be obtained by reacting an aniline-formaldehyde condensation product of the formula



where n is as previously defined, with maleic anhydride according to any conventional procedure. The resulting polymaleimide may contain polymeric by-products having one or more branched structural units of the formula

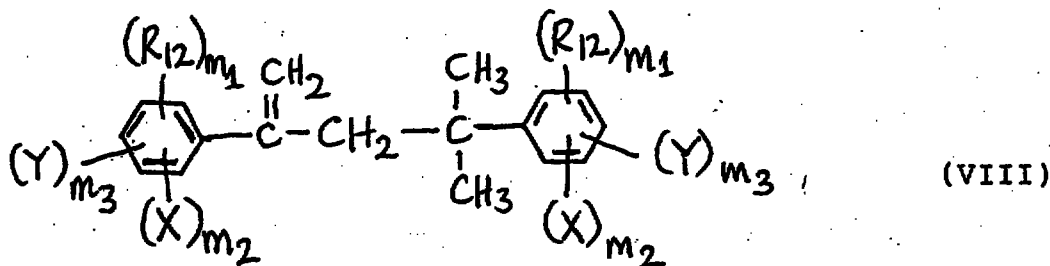


The maleimide compounds of formulas (I) and (II) may be used either alone or in combination.

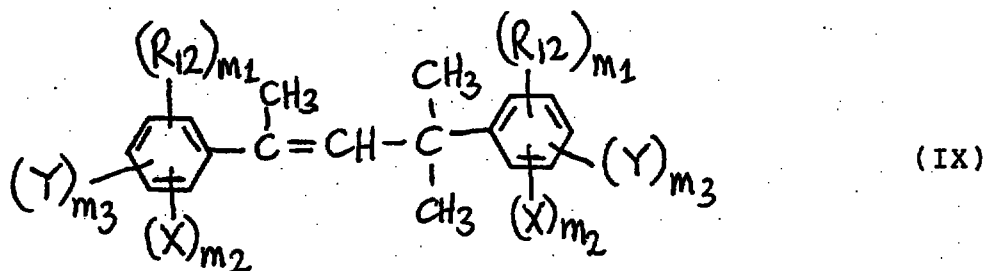
Among the alkenylphenyl compounds of formula (III) are alkenylaniline derivatives. Specific examples of the alkenylaniline derivatives include aminostyrene

(o-, m-, and p-isomers), isopropenylaniline (o-, m-, and p-isomers), isopropenyltoluidine (for example, 1,3,2-, 1,4,2-, and 1,2,3-isomers), vinyltoluidine (for example, 1,3,2-, 1,4,2-, and 1,2,3-isomers), 4-amino- α -phenylstyrene, N-(p-vinyl)maleanilic acid, N-(p-isopropenylphenyl)maleamic acid, N-(2-chloro-p-isopropenylphenyl)maleamic acid, N-(p-vinylphenyl)-maleimide, N-(p-isopropenylphenyl)maleimide, N-(p-isopropenylphenyl)-2-methylmaleimide, N-(m-isopropenylphenyl)maleimide, N-(2-methyl-p-isopropenylphenyl)maleimide, N-(p- α -ethylvinylphenyl)-maleimide, 1-vinyl-3-glycidylaminobenzene, 1-vinyl-4-glycidylaminobenzene, 1-vinyl-4-diglycidylaminobenzene, 1-isopropenyl-3-glycidylaminobenzene, 1-isopropenyl-4-glycidylaminobenzene, 1-isopropenyl-4-diglycidylaminobenzene, N-(p-isopropenyl isopropenylphenyl)isomaleimide, 1-isopropenyl-4-acetylaminobenzene, 1-isopropenyl-4-dimethylaminobenzene, and the like.

The linear dimers of the above-defined alkenylphenyl compounds can be represented by the general formulas



and



where R_{12} , X , Y , m_1 , m_2 , and m_3 are as previously defined

for formula (III). Specific examples of the linear dimers of the above-defined alkenylaniline derivatives

include 4-methyl-2,4-bis(o-aminophenyl)-1-pentene, 4-

5 methyl-2,4-bis(m-aminophenyl)-1-pentene, 4-methyl-2,4-bis-

(p-aminophenyl)-1-pentene, 4-methyl-2,4-bis(o-amino-

phenyl)-2-pentene, 4-methyl-2,4-bis(m-aminophenyl)-2-

pentene, 4-methyl-2,4-bis(p-aminophenyl)-2-pentene, 4-

methyl-2,4-bis[p-(2'-carboxyvinylcarbonylamino)phenyl]-1-

10 pentene, 4-methyl-2,4-bis[p-(2'-carboxyvinylcarbonyl-

amino)phenyl]-2-pentene, 4-methyl-2,4-bis(p-N-

maleimidophenyl)-1-pentene, 4-methyl-2,4-bis(p-N-

maleimidophenyl)-2-pentene, 4-methyl-2,4-bis(p-N-

isomaleimidophenyl)-1-pentene, 4-methyl-2,4-bis(p-N-

15 isomaleimidophenyl)-2-pentene, 4-methyl-2,4-bis(3'-

methyl-4'-N-maleimidophenyl)-1-pentene, 4-methyl-2,4-bis-

(3'-methyl-4'-N-maleimidophenyl)-2-pentene, 4-methyl-2,4-

bis(3',5'-dichloro-4'-N-maleimidophenyl)-1-pentene,

4-methyl-2,4-bis(3',5'-dichloro-4'-N-maleimidophenyl)-2-

20 pentene, and the like.

The chemical makeup of the thermosetting resin

composition of the invention may vary widely according to the purpose of its use. The weight ratio of the maleimide compound to the alkenylphenyl compound can generally range from 1:1 to 300:1, the preferred range being from 5:1 to 200:1. If the amount of maleimide compound used is below the aforesaid limits, the resin composition as used in the form of a varnish will have a shortened pot life and the cured product will show a lowering of thermal stability or heat resistance, while if it is above the aforesaid limits, the resin composition will show a decrease in curing rate and will have such poor curability that the cured product may fail to achieve sufficient mechanical strength:

The thermosetting resin composition of the invention can contain a polymerizable unsaturated compound in addition to the maleimide compound and the alkenylphenyl compound. The term "polymerizable unsaturated compound" as used herein denotes vinyl monomers, radical-polymerizable unsaturated polymers, and radical-polymerizable unsaturated oligomers. Specific examples of the vinyl monomers include styrene, substituted styrenes having their substituent or substituents on the benzene nucleus, acrylonitrile, acrylic acid and its esters, methacrylic acid and its esters, acrylamide, glycidyl acrylate, glycidyl methacrylate, divinylbenzene, divinyltoluene, diallyl phthalate, triallyl cyanurate, tribromophenyl allyl ether, and the like. Specific examples of the radical-polymerizable unsaturated

polymers and oligomers include unsaturated polyester, polybutadiene, polychloroprene, polypentadiene, and diallyl phthalate prepolymer; the foregoing polymers and oligomers modified by the addition of styrene, maleic acid, or epoxy resin; cyclic oligomers such as dicyclopentadiene and cyclododecatriene; terpenoids; and the like. These polymerizable unsaturated compounds may be used either alone or in combination.

The amount of polymerizable unsaturated compound used depends on whether it is a monomer, polymer, or oligomer. However, the ratio of the combined weight of the maleimide compound and the alkenylphenyl compound to the weight of the polymerizable unsaturated compound can generally range from 1:50 to 200:1, the preferred range being from 1:20 to 100:1. If the amount of polymerizable unsaturated compound used is above the aforesaid limits, the cured product will show a lowering of heat resistance and storage stability, while if it is below the aforesaid limits, the fluidity of the resin composition will be too low to exhibit good molding properties.

The thermosetting resin composition of the invention can be cured by application of heat. Although no particular limitation is placed on the curing temperature, the preferred range is from 30 to 300°C. It can also be cured by exposure to radiation such as ultraviolet rays, X-rays, alpha rays, beta rays, and gamma rays.

Depending on the purpose of its use, the

thermosetting resin composition of the invention can be either mixed with one or more additional components or dissolved in an organic solvent. More specifically, if the resin composition is to be used as a molding material, one or more inorganic fillers such as silica, silica glass, clay, aluminium hydroxide, asbestos, glass fiber, mica, gypsum, kaolin, cement, talc, zinc white, graphite, magnesite, molybdenum disulfide, titanium white, silica sand, carbon black, barium sulfate, and calcium carbonate can be incorporated thereinto. Moreover, a conventional flame retarder such as brominated organic compounds, antimony compounds, and phosphorus compounds can be added to the resin composition for the purpose of imparting flame-retardant properties thereto. If the resin composition is to be used in the form of a solution, it can be dissolved in an organic solvent such as N-methyl-2-pyrrolidone, dimethylformamide, dimethylacetamide, dimethyl sulfoxide, dioxane, acetone, methyl ethyl ketone, tetrahydrofuran, cellosolve (trade mark for 2-ethoxyethanol), methyl acetate, ethyl acetate, chloroform, benzene, toluene, xylene, and chlorobenzene.

The thermosetting resin composition of the invention can give a cured product having excellent heat resistance, storage stability, and moisture absorption resistance as well as good electrical and mechanical properties, and can bring about an improvement in operating efficiency owing to its good molding and fast-curing properties.

Accordingly, it has a wide variety of industrial applications and can be used, for example, in the manufacture of impregnating resins, adhesives, powder paints and coatings, and molding materials. Moreover, 5 when used in the vulcanization of rubber, it can exhibit excellent crosslinking and vulcanization-accelerating effects thereby imparting improved characteristics, such as very high resistance to heat deterioration, to the vulcanized rubber. Thus, it will be apparent to those 10 skilled in the art that the thermosetting resin composition of the invention is an exceptionally good material having a wide spectrum of functionality.

The present invention is further illustrated by the following examples. In these examples, all parts and 15 percentages are by weight unless otherwise noted.

Examples 1- 5

To 53.7 g of N,N'-(methylenedi-p-phenylene)bis-maleimide were added 10.5 g, 3.5 g, 2.5 g, 1.5 g, and 20 0.8 g, respectively, of N-(p-isopropenylphenyl)maleimide. These ingredients were thoroughly mixed to form a total of 5 resin compositions. Each resin composition was melted on an iron plate preheated to and kept at 180°C, and then stirred well with a spatula to measure the time 25 required for gelation. In addition, the resulting gel was measured by thermogravimetry to measure its loss in weight due to thermal decomposition, the loss in weight

being regarded as a measure of its thermal decomposition. The results thus obtained are summarized in Table 1.

Table 1

Example	N,N'-(Methylenedi-p-phenylene)-bismaleimide (g)	N-(p-Isopropenylphenyl)-maleimide (g)	Gelation Time (sec)	Temperature at 1% Loss in Weight (°C)
1	53.7	10.5	20	476
2	53.7	3.5	37	500
3	53.7	2.5	41	496
4	53.7	1.5	77	496
5	53.7	0.8	188	496

Examples 6 - 11

To 53.9 g of N,N'-(Methylenedi-p-phenylene)bismaleimide were added 10.0 g, 6.7 g, 3.3 g, 2.4 g, 1.3 g, and 0.5 g, respectively, of an equimolar mixture of 4-methyl-2,4-bis(p-aminophenyl)-1-pentene and 4-methyl-2,4-bis(p-aminophenyl)-2-pentene. These ingredients were thoroughly mixed to form a total of 6 resin compositions. The gelation time of each resin composition and its loss in weight due to thermal decomposition were measured in the same manner as described in Example 1. The results thus obtained are summarized in Table 2.

Table 2

Example	N,N'-(Methylenedi-p-phenylene)-bismaleimide (g)	Diamine ¹⁾ (g)	Gelation Time (sec)	Temperature at 1% Loss in Weight (°C)
6	53.7	10.0	57	418
7	53.7	6.7	74	438
8	53.7	3.3	128	460
9	53.7	2.4	134	468
10	53.7	1.3	214	478
11	53.7	0.5	299	490

1) An equimolar mixture of 4-methyl-2,4-bis(p-aminophenyl)-1-pentene and 4-methyl-2,4-bis(p-aminophenyl)-2-pentene

Example 12

To 53.7 g of N,N'-(methylenedi-p-phenylene)bismaleimide was added 2.4 g of an alkenylaniline derivative composition consisting of 7.5% of p-isopropenylaniline, 63% of 4-methyl-2,4-bis(p-aminophenyl)-1-pentene, 20.7% of 4-methyl-2,4-bis(p-aminophenyl)-2-pentene, and 8.8% of p-isopropenylaniline trimer. These ingredients were thoroughly mixed to form a resin composition. When measured in the same manner as described in Example 1, the gelation time of this resin composition was found to

be 124 seconds and the temperature at 1% loss in weight was found to be 472°C.

Comparative Example 1

5 N,N'-(Methylenedi-p-phenylene)bismaleimide alone was melted on an iron plate preheated to and kept at 180°C, and then stirred well with a spatula to measure the time required for gelation. As a result, the gelation time was found to be 1,307 seconds.

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Example 13

 An intimate mixture of 10 g of styrene, 7 g of N,N'-(methylenedi-p-phenylene)bismaleimide, and 1.5 g of N-(p-isopropenylphenyl)maleimide was prepared. Upon
15 heating at 120°C, it cured in 28 minutes. When styrene was used alone, little polymerization product was noted even after 3 hours' heating at that temperature. Moreover, a mixture of styrene and the aforesaid bismaleimide showed similar unsuccessful results.

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Example 14

 An intimate mixture of 7.0 g of a resin composition (commercially available under the trade name of ESTAR XE7472 from Mitsui Toatsu Chemicals, Inc.), 1.0 g of N,N'-
25 (methylenedi-p-phenylene)bismaleimide, and 0.2 g of N-(p-isopropenylphenyl)maleimide was prepared. The aforesaid resin composition consisted of about 55% of

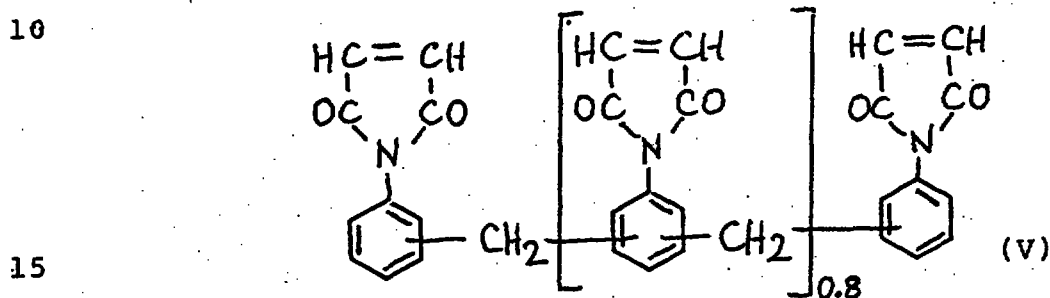
unsaturated polyester, about 45% of styrene, and 0.02% of hydroquinone. Upon heating at 120°C, this mixture cured in 13 minutes. When ESTER XE7472 was used alone, little polymerization product was noted even after 200 5 minutes' heating at that temperature.

Example 15

An intimate mixture of 10 g of liquid polybutadiene (commercially available under the trade name of NISSO-PB 10 G-1000 from Nippon Soda Company, Limited), 10 g of styrene, 7 g of N,N'-(methylenedi-p-phenylene)bis-maleimide, and 2.0 g of 4-methyl-2,4-bis(p-aminophenyl)-1-pentene was prepared. Upon heating at 120°C, it cured in 21 minutes.

Example 16

An intimate mixture of 10 g of liquid polybutadiene (commercially available under the trade name of NEW POLYOIL NX-5 from Nippon Zeon Co. Ltd.), 10 g of styrene, a quantity of an alkylphenyl compound (component (b) as hereinbefore defined), and 7 g of a polymethylene polyphenylene polymaleimide of formula (V) in which n had an average value of 0.8 was prepared. Thus, the aforesaid polymaleimide is represented by the formula



Upon heating at 120°C, this mixture cured in 27 minutes.

Example 17

An intimate mixture of 7.0 g of a resin composition (commercially available under the trade name of ESTAR XE7472 from Mitsui Toatsu Chemicals, Inc.) 1.0 g of N,N'-(methylenedi-p-phenylene)bismaleimide, and 0.2 g of an alkenylaniline derivative composition was prepared. The aforesaid resin composition consisted of about 55% of unsaturated polyester, about 45% of styrene, and 0.02% of hydroquinone, and the aforesaid alkenylaniline derivative composition consisted of 7.5% of p-isopropenylaniline, 42.0% of 4-methyl-2,4-bis(p-aminophenyl)-1-pentene, 30.2% of 4-methyl-2,4-bis(p-aminophenyl)-2-pentene, and 20.3% of p-isopropenylaniline trimer. Upon heating at 120°C, this mixture cured in 11.5 minutes.

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CLAIMS:

1. A thermosetting resin composition comprising:

(a) at least one maleimide compound selected from

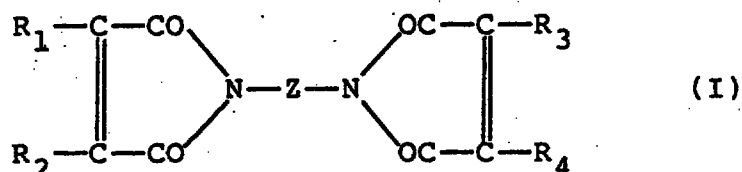
5 bismaleimides and polymaleimides; and

(b) at least one alkenylphenyl compound selected from alkenylanilines and derivatives thereof, linear dimers thereof, and polymers thereof.

10 2. A thermosetting resin composition comprising:

(a) at least one maleimide compound selected from

(i) bismaleimides having the formula



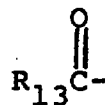
15 where R_1 , R_2 , R_3 , and R_4 are hydrogen atoms, halogen atoms; or straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals; or substituted phenyl radicals having one or more substituents selected from straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, halogen atoms, $R_{13}O-$ groups in which R_{13} is an aliphatic radical of from 1 to 5 carbon atoms, and, if there are two or more radicals represented by R_{13} , they may be identical to or different from each other,

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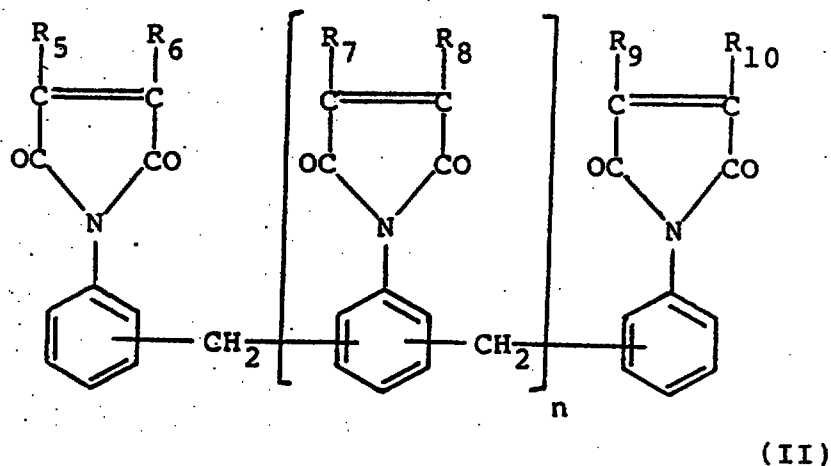
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groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups; and the radicals represented by R_1 , R_2 , R_3 , and R_4 may be identical to or different from each other, and Z is a divalent organic radical of at least 2 carbon atoms; and (ii) polymaleimides having the formula



where R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} are hydrogen atoms; halogen atoms; straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals, or substituted phenyl radicals having one or more substituents selected from halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, $R_{13}O$ -groups in which R_{13} is as previously defined,

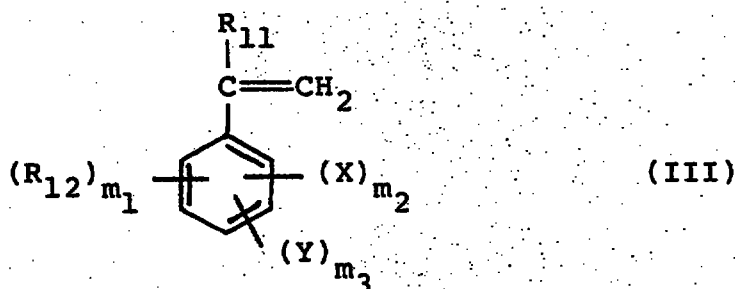


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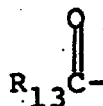


groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups, and the radicals represented by R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} may be identical to or different from each other; and n is zero or a positive number; and

(b) at least one alkenylphenyl compound selected from 10 compounds having the formula



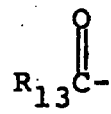
where R_{11} and R_{12} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, phenyl radicals, or substituted phenyl radicals having one or more substituents selected from halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, $\text{R}_{13}\text{O}-$ groups in which R_{13} is as previously defined,



groups in which R_{13} is as previously defined, hydroxyl groups, and cyano groups, and the radicals represented by R_{11} and R_{12} may be identical to or different from each

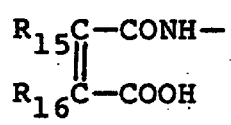


other, X is a hydrogen atom, halogen atom, carboxyl group,



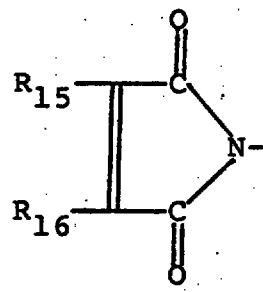
group wherein R_{13} is as previously defined, $\text{R}_{13}\text{O}-$ group

- 5 wherein R_{13} is as previously defined, or cyano group, Y is
an amino group, glycidylamino group,
diglycidylamino group,



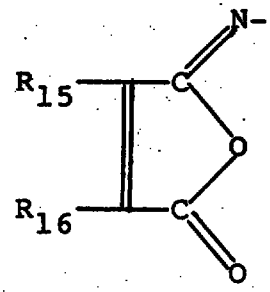
- 10 group in which R_{15} and R_{16} are hydrogen atoms, halogen
atoms, or straight-chain or branched alkyl radicals of from
1 to 10 carbon atoms and the radicals represented by R_{15} and
 R_{16} may be identical to or different from each other,

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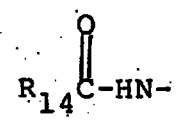


group in which R_{15} and R_{16} are as previously defined,

20



- 25 group in which R_{15} and R_{16} are as previously defined,





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group in which R_{14} is an aliphatic radical of from 1 to 20 carbon atoms and, if there are two or more radicals represented by R_{14} , they may be identical to or different from each other, alkylamino group of from 1 to 5 carbon atoms, or dialkylamino group in which each alkyl radical has from 1 to 5 carbon atoms, and m_1 , m_2 , and m_3 are positive integers and the sum of $m_1 + m_2 + m_3$ has a value of 5; linear dimers thereof; and polymers thereof.

3. A thermosetting resin composition according to claim 2 wherein, in formula (I), R_1 , R_2 , R_3 , and R_4 are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 5 carbon atoms, or phenyl radicals and the radicals represented by R_1 , R_2 , R_3 , and R_4 may be identical to or different from each other, and Z is an organic radical of 2 or more carbon atoms selected from aliphatic, alicyclic, aromatic, and heterocyclic radicals and combinations thereof or a combination of an organic radical as defined above with -O-, -S-, -S-S-, -CO-, -SO-, -SO₂-, -CO₂-, -N=N-, -NH-, or -CONH-; in formula (II), R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 5 carbon atoms, or phenyl radicals and the radicals represented by R_5 , R_6 , R_7 , R_8 , R_9 , and R_{10} may be identical to or different from each other, and n has a value of from 0.1 to 10; and in formula (III), R_{11} and R_{12} are hydrogen atoms, halogen atoms, straight-chain or branched alkyl radicals of from 1 to 10 carbon atoms, or phenyl radicals



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and the radicals represented by R_{11} and R_{12} may be identical to or different from each other, X is a hydrogen or halogen atom, Y is an amino group, maleimido group, glycidylamino group, or alkylamino group of from 1 to 5 carbon atoms, and m_1 , m_2 , and m_3 are positive integers and the sum of $m_1 + m_2 + m_3$ has a value of 5.

4. A thermosetting resin composition according to claim 2 or claim 3 wherein the maleimide compound is N,N'-10 (methylenedi-p-phenylene)bismaleimide.

5. A thermosetting resin composition according to claim 2 or claim 3 wherein the maleimide compound is a polymaleimide of formula (II) in which n has a value of from 15 0.1 to 10.

6. A thermosetting resin composition according to any one of claims 2 to 5 wherein the weight ratio of the maleimide compound to the alkenylphenyl compound ranges from 20 1:1 to 300:1.

7. A thermosetting resin composition according to any one of claims 2 to 6 wherein the alkenylphenyl compound is



selected from p-isopropenylaniline and linear dimers thereof.

8. A thermosetting resin composition according to any
5 one of claims 2 to 6 wherein the alkenylphenyl compound is selected from N-(p-isopropenylphenyl)maleimide and linear dimers thereof.

9. A thermosetting resin composition according to claim
10 1 which contains at least one polymerizable unsaturated compound in addition to the maleimide compound and the alkenylphenyl compound.

10. A thermosetting resin composition according to any
15 one of claims 2 to 8 which contains at least one polymerizable unsaturated compound in addition to the maleimide compound and the alkenylphenyl compound.

11. A thermosetting resin composition according to claim
20 10 wherein the polymerizable unsaturated compound is at least one compound selected from vinyl monomers, radical-polymerizable unsaturated polymers, and radical-polymerizable unsaturated oligomers.

25 12. A thermosetting resin composition according to claim 10 wherein the polymerizable unsaturated compound is at least one vinyl monomer selected from styrene, substituted



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styrenes, acrylonitrile, acrylic acid, acrylic esters, methacrylic acid, methacrylic esters, acrylamide, glycidyl acrylate, and glycidyl methacrylate.

- 5 13. A thermosetting resin composition according to claim 10 wherein the polymerizable unsaturated compound is unsaturated polyester, cyclized polybutadiene, or polybutadiene having the 1,2-double bond.
- 10 14. A thermosetting resin composition according to claim 10 wherein the ratio of the combined weight of the maleimide compound and the alkenylphenyl compound to the weight of the polymerizable unsaturated compound ranges from 1:50 to 200:1.
- 15
15. A thermosetting resin composition according to claim 1 or claim 2 substantially as described herein.
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