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(54) **COMPOSITION A BASE DE RESINE DE STYRENE, RENFORCEE DE CAOUTCHOUC; METHODE DE PREPARATION**

(54) **RUBBER-REINFORCED STYRENE RESIN COMPOSITION AND PROCESS FOR PRODUCING THE SAME**

(57) Disclosed is a rubber-reinforced styrene resin composition consisting essentially of: (1) 2-30% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (I) is grafted with less than 30 parts by weight of a polymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers, (2) 5-40% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (II) is grafted with 30 parts by weight or more of a polymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers, and (3) 40-93% by weight of a styrene copolymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers. The rubber-reinforced styrene resin composition has excellent matting effect and good physical property balance in respect of heat resistance, processability and impact resistance.

ABSTRACT OF THE DISCLOSURE

Disclosed is a rubber-reinforced styrene resin composition consisting essentially of:

(1) 2-30% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (I) is grafted with less than 30 parts by weight of a polymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers,

(2) 5-40% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (II) is grafted with 30 parts by weight or more of a polymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers, and

(3) 40-93% by weight of a styrene copolymer of a monomer component consisting of 10-95% by weight of an aromatic alkenyl compound, 1-60% by weight of an alkenyl cyanide compound and 0-89% by weight of other copolymerizable monomers. The rubber-reinforced styrene resin composition has excellent matting effect and good physical property balance in respect of heat resistance, processability and impact resistance.

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The present invention relates to a rubber-reinforced styrene resin composition having excellent heat resistance, impact resistance and processability, as well as to a process for producing the composition.

Rubber-reinforced thermoplastic resins have excellent impact resistance, processability and surface gloss and are, therefore, used in various applications. However, when they are used as shaped articles such as interior trims of vehicles, the articles are required to have mat surface. Further, some other uses require that the said resins have heat resistance. The conventional methods of delustering shaped articles include a method in which an oxide of magnesium, calcium or the like is added. This method, however, has a drawback that the impact strength is greatly reduced and the gloss is not uniformly taken off. There is also a delustering method in which a rubbery elastomer is added. This method, however, brings about deterioration of mechanical properties (in particular, hardness and rigidity) of resin.

There is further known a delustering method in which a resin component three-dimensionally cross-linked with a crosslinking monomer is added. This method, however, is unable to give a uniformly delustered molded article and moreover reduces the processability of the resin.

Meanwhile, for the purpose of improving heat resistance, it is known to use α-methylstyrene as a comonomer or to add a maleimide copolymer. This approach, however, brings about a great reduction in processability and impact resistance.

The present invention has solved the above-mentioned problems of the prior art by providing a rubber-reinforced styrene resin composition having an excellent wetting effect and a good balance of heat resistance, processability and impact resistance.

According to the present invention, there is provided a rubber-reinforced styrene resin composition consisting essentially of

(1) 2-30% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (I) is grafted with less than 30 parts by weight of a polymer (namely, the grafting degree being less than 30% by weight) of a monomer component (A) consisting of (a) 10-95% by weight of at least one aromatic alkenyl compound, (b) 1-60% by weight of at least one alkyl cyanide compound and (c) 0-89% by weight of at least one other copolymerizable monomer [(a) + (b) + (c) = 100% by weight],

(2) 5-40% by weight of a grafted rubbery polymer wherein 100 parts by weight of a rubbery polymer (II) is grafted with 30 parts by weight or more of a polymer (namely, the grafting degree being at least 30%

by weight) of a monomer component (A′) consisting of (a′) 10-95% by weight of at least one aromatic alkenyl compound, (b′) 1-60% by weight of at least one alkenyl cyanide compound and (c′) 0-89% by weight of at least one other copolymerizable monomer [(a′) + (b′) + (c′) = 100% by weight], and

(3) 40-93% by weight of a styrene copolymer of a monomer component (A") consisting of (a") 10-95% by weight of at least one aromatic alkenyl compound, (b") 1-60% by weight of at least one alkenyl cyanide compound and (c") 0-89% by weight of at least one other copolymerizable monomer [(a") + (b") + (c") = 100% by weight] [(1) + (2) + (3) = 100% by weight]; and a process for producing such a rubber-reinforced styrene resin composition.

The rubbery polymers (I) and (II) used in the rubber-reinforced styrene resin composition of the present invention may be the same or different, but are preferably the same. The rubbery polymers (I) and (II) include, for example, diene rubbery polymers such as polybutadiene, polyisoprene, styrene-butadiene random copolymer, acrylonitrile-butadiene copolymer, styrene-butadiene block copolymer and the like; hydrogenation products of said diene rubbery polymers; ethylene-propylene-(diene) rubbers; and acrylic rubbers. Of these, preferable are polybutadiene, styrene-butadiene copolymer, a hydrogenation product of polybutadiene, a hydrogenation product of styrene-butadiene block copolymer and an ethylene-propylene-(diene) rubber. The rubbery polymers mentioned above can be used alone or in combination of two or more as the rubbery polymers (I) or (II).

The aromatic alkenyl compounds which are the components (a), (a') and (a") in the monomer components (A), (A') and (A"), respectively, include styrene, t-butylstyrene, α-methylstyrene, p-methylstyrene, divinylbenzene, 1,1-diphenylstyrene, N,N-diethyl-p-aminoethylstyrene, N,N-diethyl-p-aminoethylstyrene, vinylpyridine, vinylxylene, monochlorostyrene, dichlorostyrene, monobromostyrene, fluorostyrene, ethylstyrene, vinylnaphthalene, etc. Styrene and α-methylstyrene are particularly preferable. These aromatic alkenyl compounds can be used alone or in combination of two or more.

The alkenyl cyanide compounds which are the components (b), (b') and (b") in the monomer components (A), (A') and (A"), respectively, include acrylonitrile, methacrylonitrile, etc. Of these, acrylonitirle is particularly preferable. These alkenyl cyanide compounds can be used alone or in combination of two or more.

The other copolymerizable monomers which are the components (c), (c') and (c") in the monomer components (A), (A') and (A"), respectively, include acrylic acid esters such as methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, amyl acrylate, hexyl acrylate, octyl acrylate, 2-ethylhexyl acrylate, cyclohexyl acrylate, dodecyl acrylate, octadecyl acrylate, phenyl acrylate, benzyl acrylate and the like; methacrylic acid esters such as methyl methacrylate, ethyl methacrylate, propyl methacrylate, butyl methacrylate, amyl methacrylate, hexyl methacrylate, octyl methacrylate, 2-ethylhexyl methacrylate, cyclohexyl methacrylate, dodecyl methacrylate, octadecyl methacrylate, phenyl methacrylate, benzyl methacrylate and the like; unsaturated dicarboxylic acid anhydrides such as maleic anhydride, itaconic anhydride, citraconic anhydride and the like, unsaturated monocarboxylic acids such as acrylic acid, methacrylic acid and the like, maleimide compounds such as maleimide, N-methylmaleimide, N-butylmaleimide, N-(p-methylphenyl)maleimide, N-phenylmaleimide, N-cyclohexylmaleimide and the like, epoxy compounds such as glycidyl methacrylate and the like; and so forth. Incidentally, the maleimide compounds include those obtained by copolymerizing styrene or the like with, for example, maleic anhydride and imidizing the resulting copolymer with aniline or the like. These copolymerizable monomers can be used alone or in combination of two or more as components (c), (c′) and (c"). Of these copolymerizable monomers, preferable are methyl methacrylate and maleimide compounds. In particular, the use of a maleimide compound is preferable because it gives a rubber-reinforced styrene resin composition well balanced in physical properties in respect of heat resistance and moldability.

The respective proportions of the aromatic alkenyl compounds (a), (a') and (a") in the monomer components (A), (A') and (A") used in the components (1) to (3) are 10-95% by weight, preferably 20-90% by weight. When the proportions are less than 10% by weight, the moldability of the resulting resin composition is inferior. When the proportions are more than 95% by weight, the heat resistance, chemical resistance and impact resistance of the resulting resin composition are inferior.

The respective proportions of the vinyl cyanide compounds (b), (b') and (b") in the monomer components (A), (A') and (A") used in the components (1) to (3) are 1-60% by weight, preferably 5-55% by weight. When the proportions are less than 1% by weight, the chemical resistance of the resulting resin composition is inferior. When the proportions are more than 60% by weight, the moldability and heat stability during molding, of the resulting resin composition are inferior.

The respective proportions of the other copolymerizable monomers (c), (c') and (c") in the monomer components (A), (A') and (A") used in the components (1) to (3) are 0-89% by weight, preferably 0-75% by weight. When the proportions are more than 89% by weight, the moldability, impact resistance and chemical resistance of the resulting resin composition are inferior.

The component (1) is a grafted rubbery polymer wherein the monomers (a), (b) and (c) are graft-polymerized directly onto the rubbery polymer (I). The grafting degree of this grafted rubbery polymer (i.e. the proportion of the graft-polymerized monomers to the rubbery polymer) is less than 30% by weight, preferably 3-28% by weight, more preferably 5-25% by weight. When the grafting degree is more than 30% by weight, no matting effect as desired is obtained.

The component (2) is a grafted rubbery polymer wherein the monomers (a'), (b') and (c') are graft-polymerized directly onto the rubbery polymer (II). The grafting degree of this grafted rubbery polymer is 30% by weight or more, preferably 35-250% by weight, more preferably 40-120% by weight. When the grafting degree is less than 30% by weight, impact resistance is unsatisfactory.

The component (3) is a styrene polymer other than the above-mentioned grafted rubbery polymers (1) and (2), and is composed of copolymers (free from rubbery polymer) obtained as by-products in the production of the components (1) and (2) and/or polymers produced separately.

The difference between the grafting degree of the component (1) and the grafting degree of the component (2) is preferably 5% by weight or more, more preferably 15% by weight or more, particularly preferably 20% by weight or more.

When the difference between the grafting degree of the component (1) and the grafting degree of the component (2) falls within the above range, the resulting resin composition has an excellent matting effect and a good physical property balance in respect of impact resistance, chemical resistance, heat stability and moldability.

The proportion of the component (1) in the rubber-reinforced styrene resin composition of the present invention is 2-30% by weight, preferably 3-25% by weight, more preferably 5-23% by weight, particularly preferably 8-21% by weight. When the proportion of the component (1) is less than 2% by weight, no sufficient matting effect is obtained. When the proportion is more than 30% by weight, the moldability is unsatisfactory.

The proportion of the component (2) in the rubber-reinforced styrene resin composition of the present invention is 5-40% by weight, preferably 7-35% by weight, more preferably 8-30% by weight, particularly preferably 10-25% by weight. When the proportion of the component (2) is less than 5% by weight, no sufficient impact resistance is obtained. When the proportion is more than 40% by weight, the moldability is unsatisfactory.

The proportion of the component (3) in the rubber-reinforced styrene resin composition of the present invention is 40-93% by weight, preferably 50-90% by weight. When the proportion of the component (3) is less than 40% by weight, the moldability is unsatisfactory. When the proportion is more than 93% by weight, the impact resistance is unsatisfactory.

The rubber-reinforced styrene resin composition of the present invention contains the components (1) to (3) as essential components. The present resin composition has a better physical property balance in respect of heat resistance, moldability and impact resistance (such a resin composition is referred to hereinafter as "preferable rubber-reinforced styrene rein" in some cases) when the component (3), i.e. the styrene polymer, is a copolymer of a monomer component (A") consisting of (a") 10-95% by weight, preferably 20-90% by weight of at least one aromatic alkenyl compound, (b") 1-60% by weight, preferably 5-55% by weight of at least one alkenyl cyanide compound, (c"-1) 0.5-50% by weight, preferably 1-40% by weight of at least one maleimide compound and (c"-2) 0-88.5% by weight, preferably 0-64.5% by weight of at least one other copolymerizable monomer [(a") + (b") + (c"-1) + (c"-2) = 100% by weight]. The maleimide compount (c"-1) may also be the maleimide component in a copolymer obtained by copolymerizing maleic anhydride with a copolymerizable monomer such as styrene or the like and imidizing the copolymer obtained with aniline or the like.

The rubber-reinforced styrene resin composition of the present invention consisting essentially of the components (1) to (3) can be produced by, for example, the typical production process mentioned below. The composition can be produced by compounding the following components (1') and (2'), the following components (1') to (3') or the following components (1') to (4'). The components (1') to (4') are compounded in such proportions that the grafting degrees, rubbery polymer contents, monomer compositions and compounding proportions of the components (1) to (3) fall within the respective ranges specified.

The rubber-reinforced styrene resin composition of the present invention is typically produced by a process comprising compounding

(1') 5-40% by weight, preferably 7-35% by weight, of a graft copolymer obtained by graft-polymerizing, in the presence of 5-80 parts by weight, preferably 10-65 parts by weight, of a rubbery polymer (I), 95-20 parts by weight, preferably 90-35 parts by weight, of a monomer component (A) consisting of (a) 10-95% by weight, preferably 20-80% by weight, of at least one aromatic alkenyl compound, (b) 1-60% by weight, preferably 5-50% by weight, of at least one alkenyl cyanide compound and (c) 0-89% by weight, preferably 0-75% by weight, of at least one other copolymerizable monomer [(a) + (b) + (c) = 100% by weight] [(I) + (A) = 100 parts by weight], the grafting degree of the grafted rubbery polymer contained in the graft copolymer being less than 30% by weight,

(2') 5-80% by weight, preferably 10-70% by weight, of a graft copolymer obtained by graft-polymerizing, in the presence of 5-80 parts by weight, preferably 10-65 parts by weight, of a rubbery polymer (II), 95-20 parts by weight, preferably 90-35 parts by weight, of the monomer component (A′) consisting of (a′) 10-95% by weight, preferably 20-80% by weight, of at least one aromatic alkenyl compound, (b′) 1-60% by weight, preferably 5-50% by weight, of at least one alkenyl cyanide compound and (c′) 0-89% by weight, preferably 0-75% by weight, of at least one other copolymerizable monomer [(a′) + (b′) + (c′) = 100% by weight] [(II) + (A) = 100 parts by weight], the grafting degree of the grafted rubbery polymer contained in the graft copolymer being 30% by weight or more,

(3′) 0-60% by weight, preferably 5-55% by weight, more preferably 10-50% by weight, of a styrene polymer obtained by copolymerizing a monomer component (A") consisting of (a") 10-95% by weight, preferably 20-80% by weight, of at least one aromatic alkenyl compound, (b") 0-60% by weight, preferably 1-50% by weight, of at least one alkenyl cyanide compound, (c"-1) 0.5-80% by weight, preferably 10-70% by weight, more preferably 15-60% by weight of at least one maleimide compound and (c"-2) 0-80% by weight, preferably 0-75% by weight, of at least one other copolymerizable monomer [(a") + (b") + (c"-1) + (c"-2) = 100% by weight], and

(4′) 0-60% by weight, preferably 0-50% by weight, of a styrene polymer obtained by copolymerizing a monomer component (A′′′) consisting of (a′′′) 10-95% by weight, preferably 20-80% by weight, of at least one aromatic alkenyl compound, (b''') 1-60% by weight, preferably 5-50% by weight, of at least one alkenyl cyanide compound and (c''') 0-89% by weight, preferably 0-75% by weight, of at least one other copolymerizable monomer [(a''') + (b''') + (c''') = 100% by weight] [(1') + (2') + (3') + (4') = 100% by weight].

The grafting degree and the content of grafted rubbery polymer in the graft copolymer are values obtained in the following manner:

One part by weight of a graft copolymer was placed in 20 parts by weight of acetone; the mixture was stirred at 25°C for 48 hours and then separated into an insoluble portion (dry weight = W) and a soluble portion (dry weight = X) using a centrifuge. Using the data obtained, the grafting degree and the grafted rubbery polymer content were determined from the following equations:

Grafting degree (% by weight) = [(W - R)/R] x 100

(R is a weight of a rubbery polymer before being subjected to grafting [i.e. rubbery polymer (I) or (II)], calculated from polymerization recipe for graft copolymer.)

Content of grafted rubbery polymer (% by weight) = [W/(W + X)] x 100

(The grafted rubbery polymer refers to the above insoluble portion.)

In the process of the present invention, the rubbery polymers (I) and (II) used in the components (1') and (2') may be respectively the same as the rubbery polymers (I) and (II) in the components (1) and (2).

Also, the monomers constituting the monomer components (A) to (A''') used in the components (1') to (4') may be the same as the monomers in the components (1) to (3). The monomer components (A) to (A''') of the components (1') to (4') may be different from one another in respect of monomer type and proportion; but the monomer components of the components (1'), (2') and (4') and/or the components (2') and (4') are preferably the same as or similar to one another in respect of monomer type and proportion and, in particular, the monomer components of the components (1') and (2') are preferably the same as or similar to each other in respect of monomer type and proportion..

The component (3) (styrene polymer) used in the preferable rubber-reinforced styrene resin and the components (3') and (4') (styrene polymers) desirably have a molecular weight distribution (the ratio of weight-average molecular weight/number-average molecular weight determined from the calibration curve of standard polystyrene obtained by a gel permeation chromatography) of preferably 3.5 or less, more preferably 3.2-1.1 and a number-average molecular weight of preferably 3×10^4 to 15×10^{14}, more preferably 5×10^4 to 12×10^4, because the resulting resin composition has an excellent physical property balance in respect of impact resistance and moldability.

The number average molecular weights of the maleimide copolymers used as the component (3) or the component (3') are preferably 3×10^4 to 15×10^4, more preferably 5×10^4 to 10×10^4.

The intrinsic viscosity [η] of the methyl ethyl ketone-soluble portion of each of the components (3), (1') and (2') is preferably 0.2 to 0.8 dl/g, more preferably 03. to 0.6 dl/g.

When these conditions are met, the physical balance in respect of imapct resistance and moldability becomes better.

The rubber-reinforced styrene resin composition of the present invention can be subjected to injection molding, extrusion molding, vacuum forming, profile molding, expansion molding, etc. to obtain automobile parts, electrical parts, household goods, various parts for industrial use, etc. In the above molding, it is possible to add conventional additives such as anti-oxidant, ultraviolet absorber, lubricant, flame retardant, antistatic agent, foaming agent, glass fiber, and other polymers such as polyamide, polycarbonate, thermoplastic polyester, polyphenylene ether, poly-vinylidene fluoride resin, diene rubber, ethylene-propylene rubber, acrylic rubber and the like.

The present invention is more specifically explained below referring to Examples. However, the Examples are merely by way of illustration and not by way of limitation.

In the Examples, parts and % are by weight unless otherwise specified.

In the Examples, measurements of properties were effected as follows.

Izod impact strength

Measured in accordance with ASTM D 256 (6 mm thickness, notched). The unit is kgf/cm^2.

Melt flow rate

Measured in accordance with ASTM D 1238 (220°C, 10 kg). The unit is g/10 min.

Heat deformation temperature

Measured in accordance with ASTM D 648 (12 mm thickness, 18.6 kgf/cm). The unit is °C.

Surface gloss (gloss value)

Measured using a gloss meter (incident light = 60°C, reflected light = 60°). The unit is %.

Reference Example 1 [Preparation of rubber-reinforced resin (1)]

Mixture (I) recipe

Polybutadiene latex (solid content)	60	parts
Styrene	11	"
Acrylonitrile	2	"
t-Dodecyl mercaptan	0.3	"
Potassium rosinate	0.25	"
Potassium hydroxide	0.005	"

Deionized water	100	"

The above mixture (I) was fed to a reactor provided with a jacket and a stirrer. The air inside the reactor was replaced with a nitrogen gas. The temperature of the contents of the reactor was elevated to 50°C while the jacket temperature was kept at 70°C. Thereto were added a solution of 0.3 part of sodium ethylenediaminetetraacetate dihydrate, 0.35 part of sodium sulfoxylate formaldehyde dihydrate and 0.01 part of ferrous sulfate in 10 parts of water and 0.1 part of cumene hydroperoxide. The resulting mixture was subjected to reaction.

After one hour from the start of the reaction, the following mixture (II) was added continuously in 2 hours, during which the reaction was continued.

Mixture (II) recipe

Styrene	19	parts
Acrylonitrile	8	"
t-Dodecyl mercaptan	0.5	"
Potassium rosinate	1.0	"
Potassium hydroxide	0.02	"
Cumene hydroperoxide	0.15	"
Deionized water	50	"

By the above polymerization was obtained a rubber-reinforced resin (a graft copolymer, the term "rubber-reinforced resin" is hereinafter used in the same meaning) [referred to hereinafter as the rubber reinforced resin (1)], as the component (1′).

Constitution of 100 parts of the rubber-reinforced resin (1):

Component (1) [rubbery polymer (I), grafting degree = 13%]	67.8 parts
Component (3) (styrene-acrylonitrile copolymer)	32.2 "

Reference Example 2 [Preparation of rubber-reinforced resin (2)]

In the presence of 55 parts of a polybutadiene, 30 parts of styrene and 15 parts of acrylonitrile were graft-polymerized in the same manner as in the production of the rubber-reinforced resin (1), to obtain a rubber-reinforced resin [referred to hereinafter as the rubber-reinforced resin (2)] as the component (2′).

Constitution of 100 parts of the rubber-reinforced resin (2):

Component (1) [rubbery polymer (I), grafting degree = 22%]	67.1 parts
Component (3) (styrene-acrylonitrile copolymer)	32.9 "

Reference Example 3 [Preparation of rubber-reinforced resin (3)]

<u>Mixture (I) recipe</u>

Polybutadiene latex (solid content)	40	parts
Styrene	14	"
Acrylonitrile	4	"
t-Dodecyl mercaptan	0.1	"
Potassium rosinate	0.25	"

Potassium hydroxide	0.005	"
Deionized water	100	"

The above mixture (I) was fed to a reactor provided with a jacket and a stirrer. The air inside the reactor was replaced with a nitrogen gas. The temperature of the contents inside the reactor was elevated to 50°C while the jacket temperature was kept at 70°C. Thereto were added a solution of 0.3 part of sodium pyrophosphate, 0.35 part of dextrose and 0.01 part of ferrous sulfate in 10 parts of water and 0.1 part of cumene hydroperoxide. The resulting mixture was subjected to reaction.

After one hour from the start of the reaction, the following mixture (II) was added continuously in 5 hours, after which the reaction was continued for 1 hour with stirring.

Mixture (II) recipe

Styrene	28	parts
Acrylonitrile	12	"
t-Dodecyl mercaptan	0.2	"
Potassium rosinate	1.0	"
Potassium hydroxide	0.02	"
Cumene hydroperoxide	0.15	"
Deionized water	50	"

By the above polymerization was obtained a rubber-reinforced resin [referred to hereinafter as the rubber-reinforced resin (3)] as the component (2′). Constitution of 100 parts of the rubber-reinforced resin (3):

Component (2) [rubbery polymer (II), grafting degree = 43%]	57.2 parts
Component (3) (styrene-acrylonitrile Copolymer)	42.8 "

Reference Example 4 [Preparation of rubber-reinforced resin (4)]

In the presence of 40 parts of a polybutadiene, 45 parts of styrene and 15 parts of acrylonitrile were graft-polymerized in the same manner as in the production of the rubber-reinforced resin (3), to obtain a rubber-reinforced resin [referred to hereinafter as the rubber-reinforced resin (4)] as the component (2'). Constitution of 100 parts of the rubber-reinforced resin (4):

Component (2) [rubbery polymer (II), grafting degree = 75%]	70.0 parts
Component (3) (styrene-acrylonitrile copolymer)	30.0 "

Reference Example 5 [Preparation of Copolymer (1)]

Into a reactor provided with a jacket and stirrer were charged 50 parts of toluene, 70 parts of styrene, 30 parts of acrylonitrile and 0.1 part of t-dedecylmercaptan, and the air inside the reactor was replaced with a nitrogen gas, after which the internal temperature was elevated to 140°C, at which temperature the mixture was subjected to reaction for 4 hours. The toluene and unreacted monomers were removed by steam stripping to recover a copolymer (a styrene copolymer) (hereinafter, the term "copolymer" is used in the same meaning) as the component (4').

Molecular weight distribution = 3.1

Number-average molecular weight = 8.5×10^4

Reference Example 6 [Preparation of copolymer (2)]

Into a reactor provided with a jacket and a stirrer were charged 100 parts of a toluene/methyl ethyl ketone (50%/50%) mixture, 50 parts of styrene, 50 parts of N-phenylmaleimide, 0.1 part of benzoyl peroxide and 0.1 part of t-dodecylmercaptan, and the air inside the reactor was replaced with a nitrogen gas, after which the internal temperature was elevated to 85°C, at which temperature the mixture was subjected to reaction for 3 hours. The toluene and unreacted monomers were removed by steam stripping to recover a copolymer as the component (3').

Molecular weight distribution = 2.9

Number-average molecular weight = 6.8×10^4

Reference Example 7 [Preparation of copolymer (3)]

The same procedure as in Reference Example 6 was repeated, except that 40 parts of styrene, 50 parts of N-phenylmaleimide and 10 parts of acrylonitrile were substituted for the 50 parts of styrene and 50 parts of N-phenylmaleimide and the t-dodecylmercaptan was not used, to obtain a copolymer as the component (3').

Molecular weight distribution = 2.7

Number-average molecular weight = 8.2×10^4

Examples 1-9 and Comparative Examples 1-8

The rubber-reinforced resins and copolymers prepared in the Reference Examples were mixed using a Henschel mixer, according to the compounding recipes shown in Tables 1 and 2. Each of the resulting mixtures was melt-mixed at a resin temperature of 230-250°C using a vented extruder and extruded therethrough to prepare pellets. The pellets were dried at 90°C. The dried pellets were subjected to injection molding at 230°C and each molded article was measured for physical properties.

The results obtained are shown in Tables 1 and 2.

Table 1

	Example								
	1	2	3	4	5	6	7	8	9
<u>Compounding recipe (parts)</u>									
Rubber-reinforced resin (1) (grafting degree = 13%) [Component (1) 67.8% + component (3) 32.2%]	20	-	30	-	-	-	-	35	-
Rubber-reinforced resin (2) (grafting degree = 22%) [Component (1) 67.1% + component (3) 32.9%]	-	20	-	15	10	40	5	-	30
Rubber-reinforced resin (3) (grafting degree = 43%) [Component (2) 57.2% + component (3) 42.8%]	30	-	-	20	-	-	40	-	40
Rubber-reinforced resin (4) (grafting degree = 75%) [Component (2) 70.0% + component (3) 30.0%]	-	30	30	25	55	43	-	10	-
Copolymer (1) [Component (4') 100%]	30	20	20	-	15	-	45	35	30
Copolymer (2) [Component (3') 100%]	20	-	-	40	20	17	10	-	-
Copolymer (3) [Component (3') 100%]	-	30	20	-	-	-	-	20	-

- to be cont'd -

Table 1 (cont'd)

Composition of rubber-reinforced styrene resin (Product) (Parts)									
Component (1)	13.6	13.4	20.3	10.1	6.7	26.8	3.4	23.7	20.1
Component (2)	17.2	21.0	21.0	28.9	38.5	30.1	22.9	7.0	22.9
Component (3)	69.2	65.6	58.7	61.0	54.8	43.1	73.7	69.3	57.0
Physical Properties									
Izod impact strength	23	18	20	16	38	33	17	15	25
Melt flow rate	11	9	14	8	5	6	18	16	19
Heat deformation temperature	106	112	107	118	101	100	97	108	91
Surface gloss value	38	41	35	44	48	29	58	33	37

Table 2

	Comparative Example							
	1	2	3	4	5	6	7	8
<u>Compounding recipe (Parts)</u>								
Rubber-reinforced resin (1)**	1.5	30	50	1.5	10	10	-	-
Rubber-reinforced resin (2)**	-	-	-	-	-	-	-	-
Polybutadiene latex (Non-grafted rubber, solid content)	-	-	-	-	-	-	20	-
Rubber-reinforced resin (3)**	30	5	10	5	-	-	-	40
Rubber-reinforced resin (4)**	-	-	-	-	70	90	30	-
Copolymer (1) [Component (4') 100%]	48.5	45	20	67	20	-	30	30
Copolymer (2) [Component (3') 100%]	20	20	20	20	-	-	-	-
Copolymer (3) [Component (3') 100%]	-	-	-	-	-	-	30	30
<u>Composition of rubber-reinforced styrene resin (Product) (parts)</u>								
Component (1)	1.0	20.3	33.9	1.0	6.8	6.8	(20)	0
Component (2)	17.2	2.9	5.7	2.9	49.0	63.0	21	22.9
Component (3)	81.8	76.8	60.4	96.1	44.2	30.2	59	77.1

- to be cont'd -

Table 2 (cont'd)

Properties								
Izod impact strength	14	7	19	5	43	49	11	15
Melt flow rate	16	19	2	30	0.3	*	16	10
Heat deformation temperature	107	106	105	109	80	78	106	113
Surface gloss value	82	33	31	84	53	48	32	88

Note: * means "impossible to measure".

**: The same as in Table 1.

As is clear from Table 1, the rubber-reinforced styrene resins of Examples 1 to 9 according to the present invention are good in matting effect and excellent in heat resistance, moldability and impact strength.

Meanwhile, as is clear from Table 2, the rubber-reinforced styrene resins of Comparative Examples 1 and 8 contain the component (1) in an amount smaller than the range specified in the present invention, and are strikingly inferior in matting effect. The rubber-reinforced styrene resin of Comparative Example 2 contains the component (2) in an amount smaller than the range specified in the present invention and is inferior in impact resistance. The rubber-reinforced styrene resin of Comparative Example 3 contains the component (1) in an amount larger than the range specified in the present invention and is inferior in moldability. The rubber-reinforced styrene resin of Comparative Example 4 contains the component (3) in an amount larger than the range specified in the present invention and is inferior in impact resistance and matting effect. The rubber-reinforced styrene resin of Comparative Example 5 contains the component (2) in an amount larger than the range specified in the present invention and is inferior in moldability. The rubber-reinforced styrene resin of Comparative Example 6 contains the component (3) in an amount larger than the range specified in the present invention and is inferior in moldability. In the resin of Comparative Example 7, the component (1) in the rubber-reinforced styrene resin of the present invention is replaced with a polybutadiene latex (a non-grafted rubber), and the resin of Comparative Example 7 is inferior in Izod impact strength.

Conventional rubber-reinforced styrene resins have had the problems that the surface gloss is not uniformly taken off and the physical property balance in respect of impact resistance, moldability and heat resistance is inferior. Meanwhile, in the rubber-reinforced styrene resin composition of the present invention, the surface gloss is uniformly taken off and the physical property balance in respect of impact resistance, moldability and heat resistance is excellent, and therefore, the resin composition has a high industrial value.

THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A rubber-reinforced styrene resin composition consisting essentially of:

(1) 2-30% by weight of a grafted rubbery polymer having a grafting degree of 5 to 25% by weight wherein 100 parts by weight of a rubbery polymer (I) is grafted with 5 to 25 parts by weight of a polymer of a monomer component (A) consisting of (a) 10-95% by weight of at least one aromatic alkenyl compound, (b) 1-60% by weight of at least one alkenyl cyanide compound and (c) 0-89% by weight of at least one other copolymerizable monomer, in which (a) + (b) + (c) = 100% by weight,

(2) 5-40% by weight of a grafted rubbery polymer having a grafting degree of 35 to 250% by weight wherein 100 parts by weight of a rubbery polymer (II) is grafted with 35 to 250 parts by weight of a polymer of a monomer component (A') consisting of (a') 10-95% by weight of at least one aromatic alkenyl compound, (b') 1-60% by weight of at least one alkenyl cyanide compound and (c') 0-89% by weight of at least one other copolymerizable monomer, in which (a') + (b') + (c') = 100% by weight, and

(3) 40-93% by weight of a styrene copolymer of a monomer component (A") consisting of (a") 10-95% by weight of at least one aromatic alkenyl compound, (b") 1-60% by weight of at least one alkenyl cyanide compound and (c") 0-89% by weight of at least one other copolymerizable monomer, in which

(a") + (b") + (c") = 100% by weight,

wherein (1) + (2) + (3) = 100% by weight,

wherein:

the rubbery copolymers (I) and (II) may be the same or different and are each a diene rubbery polymer selected from the group consisting of polybutadiene, polyisoprene, styrene-butadiene random copolymer, acrylonitrile-butadiene copolymer and styrene-butadiene block copolymer; hydrogenation product of the diene rubbery polymer; ethylene-propylene on ethylene-propylene-diene rubber; or acrylic rubber;

the aromatic alkenyl compounds (a), (a') and (a") each comprise at least one member selected from the group consisting of styrene, t-butylstyrene, α-methylstyrene, p-methylstyrene, 1,1-diphenylstyrene, vinylpyridine, vinylxylene, monochlorostyrene, dichlorostyrene, monobromostyrene, fluorostyrene, ethylstyrene, and vinylnaphthalene;

the alkenyl cyanide compound (b), (b') and (b") each comprises at least one member selected from the group consisting of acrylonitrile and methacrylonitrile; and

the other copolymerizable monomers (c), (c') and (c") each comprise at least one member selected from the group consisting of acrylic acid esters, methacrylic acid esters, unsaturated dicarboxylic acid anhydrides, unsaturated monocarboxylic acids, maleimide compounds and glycidyl methacrylate.

2. The rubber-reinforced styrene resin composition

according to Claim 1, wherein the ratio of the component (1)/the component (2)/the component (3) is 3-25/7-35/50-90% by weight.

3. The rubber-reinforced styrene resin composition according to Claim 1, wherein the ratio of the component (1)/the component (2)/the component (3) is 8-21/10-25/50-82% by weight.

4. The rubber-reinforced styrene resin composition according to Claim 1, wherein the grafting degree of the component (2) is 40-120% by weight.

5. The rubber-reinforced styrene resin composition according to Claim 2, wherein the grafting degree of the component (2) is 40-120% by weight.

6. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the difference between the grafting degree of the component (1) and the grafting degree of the component (2) is 5% by weight or more.

7. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the difference between the grafting degree of the component (1) and the grafting degree of the component (2) is 20% by weight or more.

8. The rubber-reinforced styrene resin composition

according to any one of Claims 1 to 5, wherein the ratio of the component (a)/the component (b)/the component (c) in the monomer component (A), the ratio of the component (a')/the component (b')/the component (c') in the monomer component (A') and the ratio of the component (a")/the component (b")/the component (c") in the monomer component (A") are each 20-90/5-55/0-75% by weight.

9. The rubber-reinforced styrene composition according to any one of Claims 1 to 5, wherein the component (3) is a styrene copolymer of a monomer component (A") consisting of (a") at least one of the aromatic alkenyl compounds, (b") at least one of the alkenyl cyanide compounds, (c"-1) at least one maleimide compound and (c"-2) at least one of the other copolymerizable monomers, except for the maleimide compound, and the ratio of the component (a")/the component (b")/the component (c"-1)/the component (c"-2) in the component (3) is 10-95/1-60/0.5-50/0-88.5% by weight.

10. The rubber-reinforced styrene resin composition according to claim 9, wherein the ratio of the component (a")/the component (b")/the component (c"-1)/the component (c"-2) in the component (3) is 20-90/5-55/1-40/0-74.5% by weight.

11. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the molecular weight distribution Mw/Mn of the component (3) is 1.1-3.2.

12. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the weight-average molecular weight (polystyrene-reduced Mw) of the component (3) is $3 \times 10^4 - 15 \times 10^4$.

13. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the intrinsic viscosity of the component (3) as measured at 25°C in methyl ethyl ketone is 0.2-0.8 dl/g.

14. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the intrinsic viscosity of the component (3) as measured at 25°C in methyl ethyl ketone is 0.2-0.6 dl/g.

15. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the aromatic alkenyl compound of each of the monomer components (A), (A') and (A") is at least one member selected from the group consisting of styrene, t-butylstyrene, p-methylstyrene, vinylpyridine, vinylxylene, monochlorostyrene, dichlorostyrene, monobromostyrne, fluorostyrene, ethylstyrene and vinylnaphthalene.

16. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the aromatic alkenyl compound of each of the monomer components (A), (A') and (A") is styrene.

17. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the alkenyl cyanide compound of each of the monomer components (A), (A') and (A") is acrylonitrile.

18. The rubber-reinforced resin composition according to any one of Claims 1 to 5, wherein the other copolymerizable monomer of each of the monomer components (A), (A') and (A") is selected from the group consisting of acrylic acid esters, methacrylic acid esters, unsaturated monocarboxylic acids, maleimide compounds and glycidyl methacrylate.

19. The rubber-reinforced styrene resin composition according to Claim 18, wherein the acrylic acid esters are selected from the group consisting of methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, amyl acrylate, hexyl acrylate, octyl acrylate, 2-ethylhexyl acrylate, cyclohexyl acrylate, dodecyl acrylate, octadecyl acrylate, phenyl acrylate and benzylacrylate; the methacrylate acid esters are selected from the group consisting of methyl methacrylate, ethyl methacrylate, propyl methacrylate, butyl methacrylate, amyl methacrylate, hexyl methacrylate, octyl methacrylate, 2-ethylhexyl methacrylate, cyclohexyl methacrylate, dodecyl methacrylate, octadecyl methacrylate, phenyl methacrylate and benzyl methacrylate; the unsaturated monocarboxylic acids are selected from the group consisting of acrylic acid and methacrylic acid; and the maleimide compounds are selected from the group consisting of maleimide, N-

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methylmaleimide, N-butylmaleimide, N-(p-methylphenyl) maleimide, N-phenylmaleimide and N-cyclohexylmaleimide.

20. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the other copolymerizable monomer of the monomer components (A), (A') and (A") is selected from the group consisting of methyl methacrylate and a maleimide compound.

21. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the rubbery polymers (I) and (II) are independently at least one member selected from the group consisting of diene rubbery polymers, hydrogenation products thereof, ethylene-propylene rubbers and ethylene-propylene-diene rubbers.

22. The rubber-reinforced styrene resin composition according to Claim 21, wherein the diene rubbery polymers are selected from the group consisting of polybutadiene, polyisoprene, styrene-butadiene random copolymer, acrylonitrile-butadiene copolymer and styrene-butadiene block copolymer.

23. The rubber-reinforced styrene resin composition according to any one of Claims 1 to 5, wherein the rubbery polymers (I) and (II) are independently at least one member selected from the group consisting of polybutadiene, styrene-butadiene copolymer, a hydrogenation product of polybutadiene,

a hydrogenation product of styrene-butadiene block copolymer, an ethylene-propylene rubber and an ethylene-propylene-diene rubber.

24. A process for producing a rubber reinforced styrene resin composition of Claim 1, which comprises compounding:

(1') 5-40% by weight of a graft copolymer obtained by graft-polymerizing, in the presence of 5-80 parts by weight of the rubbery polymer (I) 95-20 parts by weight of a monomer component (A) consisting of (a) 10-95% by weight of at least one of the aromatic alkenyl compounds, (b) 1-60% by weight of at least one of the alkenyl cyanide compounds and (c) 0-89% by weight of at least one of the other copolymerizable monomers, wherein (a) + (b) + (c) = 100% by weight, and (I) + (A) = 100 parts by weight, the grafting degree of the grafted rubbery polymer contained in the graft copolymer being 5 to 25% by weight,

(2') 5-80% by weight of a graft copolymer obtained by graft-polymerizing, in the presence of 5-80 parts by weight of the rubbery polymer (II), 95-20 parts by weight of a monomer component (A') consisting of (a) 10-95% by weight of at least one of the aromatic alkenyl compounds, (b) 1-60% by weight of at least one of the alkenyl cyanide compounds, and (c) 0-89% by weight of at least one of the other copolymerizable monomers, wherein (a') + (b') + (c') = 100% by weight and (II) + (A') = 100 parts by weight, the grafting degree of the grafted rubbery polymer contained in the graft copolymer being

35 to 250% by weight,

(3') 0-60% by weight of a styrene polymer obtained by copolymerizing a monomer component (A") consisting of (a") 10-95% by weight of at least one of the aromatic alkenyl compounds, (b") 0-60% by weight of at least one of the alkenyl cyanide compounds, (c"-1) 0.5-80% by weight of at least one maleimide compound and (c"-2) 0-80% by weight of at least one of the other copolymerizable monomers except for the maleimide compound, wherein (a") + (b") + (c"-1) + (c"-2) = 100% by weight, and

(4') 0-60% by weight of a styrene polymer obtained by copolymerizing of a monomer component (A''') consisting of (a''') 10-95% by weight of at least one of the aromatic alkenyl compounds, (b''') 1-60% by weight of at least one of the alkenyl cyanide compounds and (c''') 0-89% by weight of at least one of the other copolymerizable monomers, wherein (a) + (b) + (c) = 100% by weight wherein (1') + (2') + (3') + (4') = 100% by weight.

25. The process according to Claim 24, wherein the ratio of the component (1')/the component (2')/the component (3')/the component (4') is 7-35/10-70/5-55/0-50% by weight.

26. The process according to Claim 24, wherein the component (1') is a graft copolymer obtained by graft-polymerizing, in the presence of 10-65 parts by weight of the rubbery polymer (I), 90-35 parts by weight of a monomer component (A) consisting of (a) 20-80% by weight of at least

one of the aromatic alkenyl compounds, (b) 5-50% by weight of at least one of the alkenyl cyanide compounds and (c) 0-75% by weight of at least one of the other copolymerizable monomers, wherein (a) + (b) + (c) = 100% by weight and (I) + (A) = 100 parts by weight, the grafting degree of the grafted rubbery polymer contained in the graft copolymer being 5 to 25% by weight; the component (2′) is a graft copolymer obtained by graft-polymerizing, in the presence of 10-65 parts by weight of the rubbery polymer (II), 90-35 parts by weight of a monomer component (A′) consisting of (a′) 20-80% by weight of at least one of the aromatic alkenyl compounds, (b′) 5-50% by weight of at least one of the alkenyl cyanide compounds and (c′) 0-75% by weight of at least one of the other copolymerizable monomers, wherein (a′) + (b′) + (c′) = 100% by weight and (II) + (A) = 100 parts by weight, the grafting degree of the grafted rubbery polymer contained in the graft copolymer being 35-250% by weight; the component (3′) is a styrene polymer obtained by copolymerizing a monomer component (A") consisting of (a") 20-80% by weight of at least one of the aromatic alkenyl compounds, (b") 1-50% by weight of at least one of the alkenyl cyanide compounds, (c"-1) 10-70% by weight of at least one maleimide compound and (c"-2) 0-75% by weight of at least one of the other copolymerizable monomers except for the maleimide compound, wherein (a") + (b") + (c"-1) + (c"-2) = 100% by weight; and the component (4′) is a styrene polymer obtained by copolymerizing a monomer component (A‴) consisting of (a‴) 20-80% by weight of at least one of the aromatic alkenyl compounds, (b‴) 5-50% by weight of at least one of the alkenyl cyanide compounds, and (c''') 0-75% by weight of at least one of the other copolymerizable monomers, wherein (a''') + (b''') + (c''') = 100% by weight.

27. The process according to Claim 24, wherein the grafting degree in the component (2') is 40-120% by weight.

28. The process according to Claim 26, wherein the grafting degree in the component (2') is 40-120% by weight.

29. The process according to any one of Claims 24 to 28, wherein the difference between the grafting degree in the component (1') and the grafting degree in the component (2') is 5% by weight or more.

30. The process according to any one of Claims 24 to 28, wherein the difference between the grafting degree in the component (1') and the grafting degree in the component (2') is 20% by weight or more.

31. The process according to any one of Claims 24 to 28, wherein the aromatic alkenyl compound of each of the monomer components (A) to (A''') is at least one member selected from the group consisting of styrene, t-butylstyrene, p-methylstyrene, vinylpyridene, vinylxylene, monochlorostyrene, dichlorostyrene, monobromostyrene, fluorostyrene, ethylstyrene, and vinylnaphthalene.

32. The process according to any one of Claims 24 to 28, wherein the aromatic alkenyl compound of each of the monomer components (A) to (A''') is styrene.

33. The process according to any one of Claims 24 to 28, wherein the alkenyl cyanide compound of each of the monomer components (A) to (A''') is acrylonitrile.

34. The process according to any one of Claims 24 to 28, wherein the at least one other copolymerizable monomer of each of the monomer components (A) to (A''') is at least one member selected from the group consisting of acrylic acid esters, methylacrylic acid esters, unsaturated monocarboxylic acids, maleimide compounds and glycidyl methacrylate.

35. The process according to Claim 34, wherein the acrylic acid esters are selected from the group consisting of methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, amyl acrylate, hexyl acrylate, octyl acrylate, 2-ethylhexyl acrylate, cyclohexyl acrylate, dodecyl acrylate, octadecyl acrylate, phenyl acrylate and benzyl acrylate; the methacrylic acid esters are selected from the group consisting of methyl methacrylate, ethyl methacrylate, propyl methacrylate, butyl methacrylate, amyl methacrylate, hexyl methacrylate, octyl methacrylate, 2-ethylhexyl methacrylate, cyclohexyl methacrylate, dodecyl methacrylate, octadecyl methacrylate, phenyl methacrylate and benzyl methacrylate; the unsaturated monocarboxylic acids are selected from the group

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consisting of acrylic acid and methacrylic acid; and the maleimide compounds are selected from the group consisting of maleimide, N-methylmaleimide, N-butylmaleimide, N-(p-methylphenyl)maleimide, N-phenylmaleimide and N-cyclohexylmaleimide.

36. The process according to any one of Claims 24 to 28 wherein the other copolymerizable monomer of each of the monomer components (A) to (A''') is at least one member selected from the group consisting of methyl methacrylate and a maleimide compound.

37. The process according to any one of Claims 24 to 28, wherein the rubbery polymers (I) and (II) are independently at least one member selected from the group consisting of diene rubbery polymers, hydrogenation products thereof, ethylene-propylene rubbers and ethylene-propylene-diene rubbers.

38. The process according to Claim 37, wherein the diene rubbery polymers are selected from the group consisting of polybutadiene, polyisoprene, styrene-butadiene random copolymer, acrylonitrile-butadiene copolymer and styrene-butadiene block copolymer.

39. The process according to Claim 38, wherein the rubbery polymers (I) and (II) are independently at least one member selected from the group consisting of polybutadiene,

styrene-butadienecopolymer, a hydrogenation product of polybutadiene, a hydrogenation product of styrene-butadiene block copolymer, an ethylene-propylene rubber and an ethylene-propylene-diene rubber.

40. A shaped article having a mat surface and being molded of the rubber-reinforced styrene resin composition according to any one of Claims 1 to 5.

41. The shaped article according to Claim 40, which is an interior trim of a motor vehicle.

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