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(54) **Rubber compositions**

(57) A rubber composition for use in tires comprises 1—30% by weight of a non-crystallizable resinous polymer having a glass transition temperature of not less than 110°C, and 70—99% by weight of at least one rubber selected from styrene-butadiene

copolymer rubber containing not more than 60% by weight of bound styrene, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%.

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SPECIFICATION

Rubber compositions

This invention relates to rubber compositions, particularly for use in tires, obtained by adding rubbery polymer with resinous polymer having a high glass transition temperature (hereinafter abbreviated as T_g), and more particularly to rubber compositions suitable for use in tires having high driving performances, i.e. considerably improved controllability and stability during high-speed running.

Automobile tires are required to take account of various factors including safety, economic efficiency, and good ride. Lately, the requirements of safety and economic efficiency have become more severe, and hence rubber tire materials are also required to satisfy various performance requirements, including high tensile strength, high tensile stress, low-temperature dependence of tensile stress, low-temperature dependence of hardness, adhesion property, wear resistance, low heat build-up, and high skid resistance.

With the development of expressway networks, it has become increasingly important to develop tires having considerably improved controllability, mainly based on response to slight steering required during changing of lanes, avoiding dangers on straight or curved roads during high-speed running of vehicles, and stability such as cornering properties.

In order to enhance the driving performances of a tire, it is necessary to increase not only hardness (modulus of elasticity) and breaking strength, particularly hardness and breaking strength at elevated temperatures of the rubber compositions forming the tire, particularly tread rubber composition, but also hysteresis loss thereof. In particular, increasing hardness is significant in improving controllability, while increasing hysteresis loss is significant in improving stability, particularly improving gripping during cornering.

In this connection, it is difficult to obtain a rubber composition simultaneously satisfying the requirements of increasing hardness and breaking strength and of reducing hysteresis loss by conventional compounding techniques. For instance, styrene-butadiene copolymer rubber (SBR) having a higher content of bound styrene when used alone has insufficient modulus of elasticity, particularly modulus of elasticity at elevated temperature. Therefore, styrene resin or styrene-butadiene block copolymer rubber is added to such an SBR, but in this case the hardness and dynamic storage modulus are reduced due to the heat build-up during high-speed running of the tire and the controllability of the tire becomes bad and the wear resistance is considerably adversely affected, so that these additions are not acceptable for the manufacture of tires having high driving performances. Furthermore, there are usually adopted a method of increasing the amount of carbon black added and a method of increasing the amount of sulfur added. However, the former method causes reduction of breaking strength and remarkable reduction of workability (occurrence of scorching on extrusion or rolling), while the latter method reduces the hysteresis loss. Further, both the methods degrade the wear resistance.

The present inventors have made various studies with respect to rubber materials for use in tires having not only high tensile strength, tensile stress and hardness at room temperature as well as high hardness, tensile strength and modulus of elasticity at an elevated temperature but also a large hysteresis loss, and have found that particular rubber compositions containing a resinous polymer with a high T_g as a main ingredient are well suitable for the desired purpose.

The present invention provides a rubber composition comprising 1 to 30% by weight of a non-crystallizable resinous polymer having a glass transition temperature of not less than 110°C , and 70 to 99% by weight of at least one rubber selected from styrene-butadiene copolymer rubber containing not more than 60% by weight of bound styrene, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%.

In the non-crystallizable resinous polymer to be used in the invention, the glass transition temperature T_g should be not less than 110°C , while the upper limit of T_g is not particularly restricted, but in practice is about 250°C . This is because, when using a resinous polymer having T_g of less than 110°C , the requirements of high modulus of elasticity and hardness at room temperature are satisfied, but the modulus of elasticity, hardness and tensile strength at an elevated temperature are not particularly improved as compared with those of a rubber composition containing no resinous polymer.

The resinous polymer may be produced by any polymerization method, among which a method, among which a method of radical polymerization using an organic peroxide as an initiator is usually used. In particular, emulsion polymerization is preferable when blending with emulsion-polymerized styrene-butadiene copolymer rubber latex.

The resinous polymer includes homopolymers made from aromatic vinyl monomers such as α -methylstyrene, nuclear-substituted styrene, and nuclear-substituted α -methylstyrene (for example, methyl group is preferable as a nuclear substitution group), acrylonitrile, vinylpyrrolidone, and vinylcarbazole, and copolymers obtained by polymerization of two or more monomers constituting the above homopolymers or by polymerization of at least two of the above-mentioned monomers and other monomers. Among these, aromatic vinyl-series resinous polymers are preferable, typical examples of which are α -methylstyrene homopolymer, α -methylstyrene-styrene copolymer, nuclear-substituted

styrene homopolymer, nuclear-substituted α -methylstyrene homopolymer, styrene-nuclear-substituted styrene copolymer, styrene-nuclear-substituted α -methylstyrene copolymer, and copolymers thereof containing diene monomer. As the diene monomer, mention may be made of butadiene, isoprene, cyclopentadiene, dicyclopentadiene, ethylidene norbornene, and 1,4-hexadiene. The diene monomer-containing polymers include, for example, α -methylstyrene-styrene-butadiene terpolymer, α -methylstyrene-styrene-isoprene terpolymer, and α -methylstyrene-isoprene copolymer. In the latter case, at least one diene monomer is added to the polymer so as not to give a glass transition temperature T_g of less than 110°C.

Among the aromatic vinyl-series resinous polymers, α -methylstyrene homopolymer, α -methylstyrene-styrene copolymer, α -methylstyrene-butadiene copolymer, α -methylstyrene-isoprene copolymer, α -methylstyrene-styrene-butadiene terpolymer and α -methylstyrene-styrene-isoprene terpolymer, each of which have a glass transition temperature T_g of not less than 110°C, are preferable in view of the balance of difficulty of availability of monomer, reactivity and T_g of the resulting polymer.

The rubber to be used in the invention is at least one rubber selected from styrene-butadiene copolymer rubber containing not more than 60% by weight of bound styrene and suitably produced by emulsion polymerization or solution polymerization using an organic alkali metal catalyst, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80% and suitably produced by solution polymerization of butadiene using a transition metal catalyst or an organic alkali metal catalyst, polybutadiene rubber having a 1,2-configuration of not less than 50% and suitably produced by solution polymerization of butadiene using an organic alkali metal catalyst, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90% and suitably produced by solution polymerization of isoprene using a transition metal catalyst or an organic alkali metal catalyst.

The amount of the rubber used is 70 to 99% by weight. When the amount is less than 70% by weight, the tensile strength, tensile stress and hardness at an elevated temperature are not improved, while when the amount exceeds 99% by weight, not only the modulus of elasticity and hardness at room temperature but also the tensile strength, modulus of elasticity and hardness at an elevated temperature are not improved.

According to a preferred embodiment of the invention, a rubber composition for use in tires comprises 1 to 30% by weight of the non-crystallizable resinous polymer and 15 to 99% by weight of styrene-butadiene copolymer rubber containing more than 30% by weight but not more than 50% by weight of bound styrene, and further contains 0 to 84% by weight of at least one rubber selected from styrene-butadiene copolymer rubber containing not more than 30% by weight of bound styrene, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%, which exhibits excellent characteristics of high hardness, high modulus of elasticity and large hysteresis loss and can considerably improve the cornering stability and high-speed stability of the tire.

According to another preferred embodiment of the invention, a rubber composition for use in tires comprises 1 to 30% by weight of the non-crystallizable resinous polymer and 30 to 99% by weight of styrene-butadiene copolymer rubber containing 15 to 30% by weight of bound styrene, and further contains 0 to 69% by weight of at least one rubber selected from natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%, which improves not only the cornering stability and high-speed stability of the tire but also the wear resistance, so that it is advantageous for applications requiring high wear resistance.

To the rubber composition according to the invention may be suitably added additives usually used in the rubber industry such as carbon black, process oil, sulfur, vulcanization accelerators, accelerator activators, and antioxidants.

The working and vulcanizing methods of the thus obtained rubber composition are not particularly restricted. For instance, ingredients for the rubber composition may be thoroughly mixed in a mixing machine such as rolls or a Banbury mixer and then vulcanized in a conventional manner using a vulcanizing pan or a vulcanizing press.

The rubber compositions according to the invention are preferably used as a tread rubber and a sidewall rubber in radial tires, particularly radial tires for passenger cars and radial tires for heavy vehicles such as trucks and buses as well as tires for motorcycles. However, the rubber compositions may also be applied not only to other tire parts such as bead portions but also to tires for construction vehicles, tires for agricultural vehicles, and bias tires for heavy vehicles such as truck and buses.

The invention will be further described with reference to the following illustrative Examples.

In these examples, a rubber composition was prepared according to a compounding recipe as shown in the following Table 1.

TABLE 1

	part by weight
Resinous polymer	10
Styrene-butadiene copolymer rubber *1	90
Zinc white	3
Stearic acid	1.5
Sulfur	2
Carbon black HAF	50
Vulcanization accelerator (Nocceler-NS) *2	0.9

*1: emulsion-polymerized styrene-butadiene copolymer rubber containing 23.5% by weight of bound styrene

*2: N-t-butyl-2-benzothiazole sulfenamide, made by Ohuchi Shinko Kagaku K.K.

EXAMPLES 1 TO 2, COMPARATIVE EXAMPLES 1 TO 2

In 200 parts by weight of water was dissolved 5 parts by weight of potassium oleate, to which was added 100 parts by weight of α -methylstyrene. To the resulting emulsion were added an activator containing 0.1 part by weight of tetrasodium ethylenediaminetetraacetate, 0.02 part by weight of ferrous sulfate heptahydrate and 0.1 part by weight of sodium dimethylsulfoxylate and further 0.1 part by weight of cumene hydroperoxide to conduct emulsion polymerization at 15°C. After 30 hours, α -methylstyrene homopolymer (hereinafter abbreviated as P α MS) was obtained in a yield of 27%. After the unreacted monomer was removed by steam distillation, the resulting homopolymer was solidified with an acid, subjected to a centrifugal dehydration and then dried in draft. The thus obtained α -methylstyrene homopolymer had Tg of 180°C as measured by a differential scanning calorimeter (DSC).

The polymerization of acrylonitrile was performed by the same method as described above, after 10 hours of which the conversion was 90%. The resulting acrylonitrile homopolymer (hereinafter abbreviated as PAN) after the drying had Tg of 125°C.

Then, a rubber composition was prepared by using the α -methylstyrene homopolymer and acrylonitrile homopolymer according to the compounding recipe shown in Table 1 and vulcanized at 145°C for 35 minutes. The properties of the resulting vulcanizate were measured to obtain results as shown in the following Table 2. For comparison, the properties of vulcanizate containing no resinous polymer (Comparative Example 1) and vulcanizate containing emulsion-polymerized polystyrene (hereinafter abbreviated as PS) with Tg of 100°C (Comparative Example 2) are also shown in Table 2.

TABLE 2

	Comparative Example 1	Example 1	Example 2	Comparative Example 2
Properties of vulcanizate				
Kind of resinous polymer	—	PαMS	PAN	PS
T _g (°C)	—	180	125	100
Addition amount of resinous polymer (part by weight)	—	10	10	10
Mooney viscosity of rubber composition ML ₁₊₄ (100°C)	50	65	59	57
200% tensile stress, kg/cm²				
25°C	104	170	165	170
70°C	96	155	142	130
100°C	95	148	131	114
120°C	94	144	125	103
300% tensile stress, kg/cm²				
25°C	205	275	265	270
70°C	185	237	222	217
100°C	176	218	202	190
120°C	171	207	187	174
Tensile strength, kg/cm²				
25°C	295	285	290	290
70°C	196	218	208	205
100°C	154	183	175	160
120°C	130	160	149	135
Hardness (JIS K—6301 spring hardness test)				
25°C	66	77	74	76
70°C	64	74	71	72
100°C	63	72	69	69
120°C	63	71	69	66

As apparent from the data of Table 2, the rubber compositions of Examples 1 and 2 each containing the non-crystallizable resinous polymer with high Tg considerably improve the modulus of elasticity (i.e. tensile stress) and hardness at room temperature and further improve the hardness, tensile strength and modulus of elasticity at elevated temperatures as compared with the rubber composition of Comparative Example 1 containing no resinous polymer.

Furthermore, it is apparent from the comparison of Comparative Example 2 with Examples 1 and 2 that when using the resinous polymer with Tg of 100°C, the hardness, tensile strength and modulus of elasticity (tensile stress) at elevated temperatures are hardly improved.

EXAMPLES 3 TO 5

In this example, α -methylstyrene and styrene were copolymerized by the same emulsion polymerization method as described in Example 1 to obtain α -methylstyrene-styrene copolymer [hereinafter abbreviated as P(α MS/S)]. That is, α -methylstyrene-styrene copolymer having Tg of 114°C was obtained in a conversion of 90% after the copolymerization at 15°C for 10 hours by using 35 parts by weight of α -methylstyrene and 65 parts by weight of styrene (Example 3). Further, α -methylstyrene-styrene copolymer having Tg of 129°C was obtained in a conversion of 60% after the copolymerization at 15°C for 20 hours by using 55 parts by weight of α -methylstyrene and 45 parts by weight of styrene (Example 4). Also, α -methylstyrene-styrene copolymer having Tg of 149°C was obtained in a conversion of 40% after the copolymerization at 15°C for 30 hours by using 85 parts by weight of α -methylstyrene and 15 parts by weight of styrene (Example 5).

Then, a latex of the α -methylstyrene-styrene copolymer was mixed with a latex of an emulsion-polymerized styrene-butadiene copolymer rubber containing 23.5% by weight of bound styrene in the mixing ratio of resinous polymer to styrene-butadiene copolymer rubber as shown in Table 1, solidified with acid-salt, dehydrated and dried in draft to form a rubber blend. This rubber blend was compounded with other ingredients according to the compounding recipe shown in Table 1 and then vulcanized in the same manner as described in Example 1. Thereafter, the properties of the resulting vulcanizate were measured to obtain results as shown in the following Table 3.

TABLE 3

	Comparative Example 2	Example 3	Example 4	Example 5	Example 1
Properties of vulcanizate					
Kind of resinous polymer	PS	P(α MS/S)	P(α MS/S)	P(α MS/s)	P α MS
Tg (°C)	100	114	129	148	180
Addition amount of resinous polymer (part by weight)	10	10	10	10	10
Mooney viscosity of rubber composition, ML ₁₊₄ (100°C)	57	58	60	62	65
200% tensile stress, kg/cm²					
25°C	170	172	169	169	170
70°C	130	138	145	150	155
100°C	114	126	133	141	148
120°C	103	113	126	138	144
300% tensile stress, kg/cm²					
25°C	270	275	274	273	275
70°C	217	220	220	232	237
100°C	190	200	205	211	218
120°C	174	180	189	203	207
Tensile strength, kg/cm²					
25°C	290	285	290	287	285
70°C	205	208	210	215	218
100°C	160	165	168	174	183
120°C	135	140	143	150	160
Hardness (JIS K—6301 spring hardness test)					
25°C	76	76	77	77	77
70°C	72	73	73	74	74
100°C	69	70	71	72	72
120°C	66	68	69	70	71

As apparent from the comparison of Examples 1 and 3 to 5 with Comparative Example 2 in Table 3, when Tg of the α -methylstyrene-styrene copolymer is not less than 110°C, the tensile strength, modulus of elasticity (tensile stress) and hardness at elevated temperatures are improved.

5 As apparent from the above, the rubber compositions having a high glass transition temperature according to the invention give improved tensile strength and hardness at room temperature as well as improved tensile strength, modulus of elasticity (tensile stress) and hardness at elevated temperatures. 5

EXAMPLES 6 TO 7, COMPARATIVE EXAMPLES 3 TO 4

10 Various vulcanizates were produced under the same conditions as described in Example 1, only changing the mixing ratio of resinous polymer to styrene-butadiene copolymer rubber in the compounding recipe of Table 1, and their properties were measured to obtain results as shown in the following Table 4. 10

TABLE 4

	Comparative Example 3	Example 6	Example 1	Example 7	Comparative Example 4
Properties of vulcanizate					
Kind of resinous polymer	P α MS	P α MS	P α MS	P α MS	P α MS
T _g (°C)	180	180	180	180	180
Addition amount of resinous polymer (part by weight)	0.5	2	10	25	35
Mooney viscosity of rubber composition, ML ₁₊₄ (100°C)	50	53	65	83	96
200% tensile stress, kg/cm²					
25°C	108	135	170	208	226
70°C	100	126	155	172	176
100°C	99	123	148	161	147
120°C	97	122	144	145	126
300% tensile stress, kg/cm²					
25°C	208	235	275	342	378
70°C	189	212	237	265	279
100°C	180	201	218	241	234
120°C	173	196	207	227	204
Tensile strength, kg/cm²					
25°C	296	290	285	257	210
70°C	195	205	218	200	172
100°C	153	165	183	176	152
120°C	130	142	160	158	138
Hardness (JIS K—6301 spring hardness test)					
25°C	66	69	77	85	90
70°C	64	68	74	75	74
100°C	64	66	72	71	66
120°C	63	65	71	69	63

As apparent from the comparison of Examples 1, 6 and 7 with Comparative Example 3, when the mixing ratio of the resinous polymer is less than 1.0% by weight, the advantages of the invention cannot be obtained. On the other hand, it is apparent from Comparative Example 4 that when the mixing ratio of the resinous polymer exceeds 30% by weight, the tensile strength lowers. From these results it is apparent that the mixing ratio of the resinous polymer is preferably within a range of 1 to 30% by weight.

EXAMPLE 8

Two rubber blends (A) and (B) were prepared in the same manner as described in Example 3 by using an α -methylstyrene-styrene copolymer having T_g of 129°C, which was obtained by copolymerizing 55 parts by weight of α -methylstyrene and 45 parts by weight of styrene at 15°C for 20 hours in the same manner as described in Example 3.

In the rubber blend (A), the mixing ratio (by weight) of α -methylstyrene-styrene copolymer to emulsion-polymerized styrene-butadiene copolymer rubber (SBR) containing 23.5% by weight of bound styrene was 15:85. In the rubber blend (B), the mixing ratio (by weight) of α -methylstyrene-styrene copolymer to emulsion-polymerized styrene-butadiene copolymer rubber (SBR) containing 35.0% by weight of bound styrene was 15:85.

Seventeen tread rubber compositions for use in radial tires for passenger cars were prepared by using the rubber blends (A) and (B) according to a compounding recipe as shown in the following Table 5. In Comparative Examples of Table 5, there was used a rubber composition containing high styrene content styrene-butadiene copolymer rubber, styrene-butadiene block copolymer rubber (content of bound styrene: 48% by weight), styrene resin, or increased amount of carbon black or sulfur. Then, seventeen radial tires for passenger cars having a tire size of 185/70 HR 14 were manufactured by using the above seventeen tread rubber compositions as a tire tread.

The hardness, maximum tensile elongation, tensile strength, dynamic storage modulus (E') and loss tangent (tan δ) of the tread rubber composition and the controllability, stability (particularly cornering stability), wear resistance and resistance to crack chipping of the tire were evaluated to obtain results as shown in Table 5.

The properties of the tire were evaluated as follows.

Controllability

The test tire was run on concrete road, on which pylons were set at an interval of 50 m, at a speed of 120 km/hr under standard internal pressure and load over a predetermined distance, during which the controllability was evaluated by a driver's feeling and defined by an index on the basis that the tire of the tread rubber composition No. 10 is 100. The larger the index value, the better the property.

Stability

The test tire was mounted on a vehicle and actually run on a circuit course under standard internal pressure and load three laps to measure an average lap time, from which an index of the stability was calculated according to the following equation:

$$\frac{\text{Average lap time of the tire of the tread rubber composition No. 10}}{\text{Average lap time of test tire}} \times 100$$

The larger the index value, the better the property.

Wear resistance

Apart from the tires used for the evaluations of controllability and stability, a tire having a two-part tread was manufactured by using the tread rubber composition No. 10 as the one part and using each of the other tread rubber compositions to be tested as the other part and then run on paved road over a distance of 50,000 km. Thereafter, the depths of the remaining tread grooves were measured, from which an index of wear resistance was calculated according to the following equation:

$$\frac{\text{Remaining groove depth of tread rubber composition to be tested}}{\text{Remaining groove depth of tread rubber composition No. 10}} \times 100$$

The larger the index value, the better the property.

Resistance to crack chipping

The test tire having the same two-part tread as used in the evaluation of wear resistance was run on a bad road consisting of 70% of unpaved road and 30% of paved road over a distance of 50,000 km. Thereafter, the appearance of the tread was visually observed on the basis of the tread rubber composition No. 10 and evaluated at three stages o, Δ and x wherein o is good, x is bad and Δ is slightly bad. 5

TABLE 5a

Tread rubber composition No.	Example											Comparative example						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	
Rubber blend (A)	100	70	-	-	-	-	-	-	-	-	-	-	-	-	-	5	-	
Rubber blend (B)	-	-	60	40	40	40	40	60	100	-	-	-	-	-	-	-	5	
Emulsion-polymerized SBR (content of bound styrene 23.5% by weight)	-	20	-	-	20	-	-	40	-	100	100	-	-	-	100	65	65	
Emulsion-polymerized SBR (content of bound styrene 40% by weight)	-	-	-	-	-	20	-	-	-	-	-	100	-	-	-	-	-	
Styrene-butadiene block copolymer rubber (content of bound styrene 48% by weight)	-	-	-	-	-	-	-	-	-	-	-	-	-	100	-	-	-	
Natural rubber	-	-	40	40	40	40	40	-	-	-	-	-	-	-	-	-	-	
Polybutadiene rubber *1	-	10	-	20	-	-	-	-	-	-	-	-	-	-	-	30	30	
Halogenated butyl rubber	-	-	-	-	-	-	20	-	-	-	-	-	-	-	-	-	-	
Compounding recipe (parts by weight)																		

TABLE 5C

Tread rubber composition No.	Example										Comparative example						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Mooney viscosity (ML ₁₊₄ , 130°C)	56	57	54	59	56	58	57	59	58	45	79	53	52	42	59	42	43
Hardness (room temperature) JIS K-6301 spring hardness test)	69	69	73	70	72	72	70	70	71	58	67	64	62	75	69	59	69
Maximum tensile elongation (room temperature) (%)	470	460	480	470	478	469	467	410	475	490	350	370	470	540	370	485	487
Tensile strength (room temperature) (kg/cm ²)	210	205	204	195	197	194	185	178	200	180	152	154	176	190	140	176	174
Hardness (100°C)	57	57	64	60	62	61	60	63	61	50	57	55	52	52	53	51	51
Maximum tensile elongation (100°C)	410	403	450	440	450	448	430	380	420	340	240	230	350	480	235	332	328
Dynamic storage modulus E' (kg/cm ²) *6	124	121	170	150	165	164	148	167	166	60	118	90	64	65	71	63	64
Loss tangent tan δ *6	0.35	0.34	0.42	0.39	0.41	0.44	0.42	0.43	0.41	0.24	0.45	0.13	0.30	0.45	0.41	0.23	0.24

TABLE 5d

Tread rubber composition No.	Example										Comparative example						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Controllability	125	123	160	140	152	150	140	158	154	100	117	84	101	102	104	100	100
Stability (cornering properties)	123	121	154	145	148	156	157	152	149	100	115	76	118	112	102	97	98
Wear resistance	125	127	105	110	108	105	103	104	107	100	43	35	60	40	54	105	104
Resistance to crack chipping	○	○	○	○	○	○	○	○	○	○	×	×	△	×	×	○	○

*1: BR-01 made by JAPAN SYNTHETIC RUBBER CO., LTD.

*2: N-phenyl-N'-isopropyl-p-phenylenediamine

*3: Diphenyl guanidine.

*4: Dibenzothiazyl disulfide .

*5: N-oxydiphenylene-2-benzothiazolesulfeneamide.

*6: Measured at 60°C and 50 Hz under static strain of 5% or dynamic strain of 1% according to tensile method.

As apparent from the data of Table 5, the controllability and stability of the tire can be greatly improved without lowering the wear resistance and resistance to crack chipping by applying the tread rubber composition according to the invention to the tread of the tire.

CLAIMS

- 5 1. A rubber composition comprising 1 to 30% by weight of a non-crystallizable resinous polymer having a glass transition temperature of not less than 110°C, and 70 to 99% by weight of at least one rubber selected from styrene-butadiene copolymer rubber containing not more than 60% by weight of bound styrene, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%. 5
- 10 2. A rubber composition as claimed in Claim 1, wherein the said resinous polymer is at least one polymer selected from α -methylstyrene homopolymer, α -methylstyrene-styrene copolymer, nuclear-substituted styrene homopolymer, nuclear-substituted α -methylstyrene homopolymer, styrene-nuclear-substituted styrene copolymer, styrene-nuclear-substituted α -methylstyrene copolymer and copolymers thereof containing diene monomer. 10
- 15 3. A rubber composition as claimed in Claim 1 or 2, which comprises 1 to 30% by weight of the said non-crystallizable resinous polymer, 15 to 99% by weight of styrene-butadiene copolymer rubber containing more than 30% by weight but not more than 50% by weight of bound styrene, and 0 to 84% by weight of at least one rubber selected from styrene-butadiene copolymer rubber containing not more than 30% by weight of bound styrene, natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%. 20
- 25 4. A rubber composition as claimed in Claim 1 or 2, which comprises 1 to 30% by weight of the said non-crystallizable resinous polymer, 30 to 99% by weight of styrene-butadiene copolymer rubber containing 15 to 30% by weight of bound styrene, and 0 to 69% by weight of at least one rubber selected from natural rubber, polybutadiene rubber having a 1,4-configuration of not less than 80%, polybutadiene rubber having a 1,2-configuration of not less than 50%, halogenated butyl rubber, butyl rubber, and polyisoprene rubber having a cis-1,4-configuration of not less than 90%. 25
- 30 5. A rubber composition according to Claim 1, substantially as herein described in any of the foregoing Examples. 30