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(54) Titre : MELANGES DE CAOUTCHOUC CONTENANT DE L'ACIDE SILIQUE, DU NOIR DE CARBONE ET DU GEL DE CAOUTCHOUC
(54) Title: RUBBER MIXTURES CONTAINING SILICIC ACID, SOOT AND RUBBER GEL

(57) Abrégé/Abstract:
The invention relates to rubber mixtures based on silicic acid and styrene/butadiene rubber solutions filled with soot, wherein the silicic acid is partially substituted by rubber gels. The vulcanised rubber produced from the inventive rubber mixtures is characterised in that has an improved hysteresis quality at 0 °C and 60 °C, and at a result an improved anti-slip resistance when wet and an improved resistance to rolling. Furthermore, said vulcanised rubber has a good resistance to abrasion. The processing quality of the unvulcanised rubber mixtures (compound viscosity and Mooney relaxation) does not deteriorate as a result of partial silicic acid substitution. Said inventive rubber mixtures are used for producing tyres and tyre components of various types and for producing technical rubber articles.
Rubber mixtures containing silica, carbon black and rubber gel

Abstract

The invention relates to rubber mixtures based on silica and carbon black-filled solution styrene/butadiene rubbers in which the silica has been partially replaced by rubber gels.

The vulcanisates produced from the rubber mixtures according to the invention are distinguished by improved cushioning behaviour at 0 °C and 60 °C, and this is accompanied by better resistance to slipping when wet and improved rolling resistance. In addition, they have good resistance to wear. The processing behaviour of the unvulcanised rubber mixtures (compound viscosity and Mooney relaxation) are not impaired by the partial substitution of silica.

The rubber mixtures according to the invention are suitable for producing an immense variety of tyres and tyre components and for producing industrial rubber articles.
Rubber mixtures containing silica, carbon black and rubber gel

The invention relates to rubber mixtures containing carbon black, silica and rubber gel and based on solution styrene/butadiene rubbers. The rubber mixtures according to the invention, in which a proportion of the silica or of the carbon black has been replaced by rubber gel are distinguished by improved cushioning behaviour at 0 °C (resistance to slipping when wet) and improved rolling resistance (improved cushioning behaviour at 60 °C), without other important mechanical properties such as the resistance to wear of the vulcanisates produced from the rubber mixtures according to the invention being impaired. Furthermore, the processing behaviour of the unvulcanised mixtures remains virtually unimpaired, as shown by the relevant characteristics (compound viscosity and Mooney relaxation) of the mixtures.

The rubber mixtures according to the invention are used, in particular, for producing tyre components of any type and for producing industrial rubber articles.

It is known to produce tyres or tyre components such as tyre treads based on silica and carbon black-filled solution styrene/butadiene rubbers (L-SBR), polybutadiene rubbers (BR) and polyisoprene rubbers (IR and NR) with a proportion of L-SBR which is greater than the proportion of other known rubbers. The vulcanisates produced from these mixtures are distinguished, in particular, by an advantageous combination of resistance to slipping when wet, rolling resistance and resistance to wear. Disadvantages when using these silica-filled rubber mixtures reside, in particular, in the high costs of specific components required such as the filler activators and the high specific weight of the silica-filled mixtures associated with processing of the rubber mixtures which is expensive in terms of energy owing to the high viscosities and the high Mooney relaxation.

It is also known to use rubber and microgels as fillers in rubber mixtures. In this connection, reference is made, in particular, to EP-A-405 216, US-A-5 395 891, US-A-6127488 and DE-A 19 701 487. Although the use of an immense variety of rubber gels as fillers in various rubber mixtures is described in the aforementioned...
patent applications and patents, the aforementioned patent applications and patents do not teach that improved cushioning behaviour at 0 °C (resistance to slipping when wet) and improved rolling resistance (improved cushioning behaviour at 60 °C) associated with low wear without impairing the processing behaviour of the unvulcanised rubber mixtures can be achieved by using small quantities of rubber gels in L-SBR rubber mixtures which contain silica and carbon black.

It was accordingly the object of the present invention to improve the aforementioned properties of rubber mixtures based on L-SBR together with good processing behaviour of the unvulcanised rubber mixtures.

The present invention accordingly relates to rubber mixtures containing carbon black, silica and rubber gel in which the above-described object is achieved.

The rubber mixtures according to the invention consist of

a) solution styrene/butadiene rubber (L-SBR)
b) silica
c) carbon black and
d) rubber gel,

wherein component a) is present in the mixture in a quantity of 100 parts by weight, component b) in quantities of 20 to 120 parts by weight, component c) in quantities of 5 to 60 parts by weight and component d) in quantities of 0.1 to 10 parts by weight, providing that the proportion of component c) is 50 wt.% maximum based on the proportion of component b).

Preferably, component a) is present in the mixture in a quantity of 100 parts by weight, component b) in quantities of 30 to 100 parts by weight, component c) in quantities of 10 to 40 parts by weight and component d) in quantities of 1.0 to 5.0 parts by weight in the mixture. The proportion of component c) is preferably 5 to 30 % based on the proportion of component b) in the mixture.
The rubber mixture according to the invention can additionally contain further known rubbers. The rubber mixture according to the invention preferably contains polybutadiene rubber (BR) and polyisoprene rubber (IR and NR) both individually and in a combination with one another in addition to the L-SBR rubbers. The proportion of polybutadiene rubber, the proportion of polyisoprene rubber or the proportion of a mixture of polyisoprene rubber and polybutadiene rubber in the mixture is 1 to 50 wt.%, preferably 5 to 40 wt.%, based on the total quantity of L-SBR rubbers present, 5 to 95 wt.% of polybutadiene rubber, based on the total mixture of polybutadiene rubber and polyisoprene rubber, being present when using a mixture of polybutadiene rubber and polyisoprene rubber.

As further component (component e)), the rubber mixtures according to the invention can also contain filler activators based on organosilicon sulphur compounds in quantities of 0.1 to 15 parts by weight, preferably 0.2 to 10 parts by weight.

In addition, the rubber mixtures according to the invention can also contain the known and conventional rubber auxiliaries and additives as well as crosslinking agents and vulcanisation accelerators (component g)) in quantities of 0.05 to 80 parts by weight, preferably 0.1 to 50 parts by weight.


Vinyl aromatic monomers suitable for making up the L-SBR rubbers include, for example, styrene, o-, m- and p-methylstyrrene, industrial methylstyrene mixtures, p-
tert.butylstyrene, α-methylstyrene, p-methoxystyrene, vinylnaphthalene, divinylbenzene, trivinylbenzene and divinylnaphthalene. Styrene is preferably used.

The content of vinyl aromatics polymerised into the L-SBR rubbers is in the range of about 5 to 50 wt.% preferably 10 to 40 wt.%.

5

Conjugate dienes suitable for making up the L-SBR rubbers include, for example: 1,3-butadiene, isoprene, 1,3-pentadiene, 2,3-dimethylbutadiene, 1-phenyl-1,3-butadiene and 1,3-hexadiene. 1,3-butadiene and isoprene, in particular butadiene are preferably used. The content of dienes polymerised into the L-SBR rubbers is in the range of about 50 to 95 wt.%, preferably in the range of 60 to 90 wt.%. The content of vinyl groups in the polymerised-in dienes lies in the range of 10 to 90 %, preferably 20 to 80 %, the content of 1,4-trans double bonds is the range of about 5 to 80 %, preferably 10 to 70 % and the content of 1,4-cis double bonds is complementary to the sum of the content of vinyl groups and 1,4-trans double bonds.

10

In the L-SBR rubbers used according to the invention, the polymerised-in monomers are usually randomly distributed. It is obviously also possible to use L-SBR rubbers with a block-like structure, which are known as integral rubbers and are described, for example, by K.-H. Nordsiek, K.-H. Kiepert in GAK Kautschuk Gummi Kunststoffe 33 (1980), No. 4, 251-255. L-SBR rubbers having a linear or branched structure or terminal groups modified in a known manner may also be used. Rubbers of this type are described, for example, in FR-A 2 053 786 and in Japanese Patent Application 56-104 906.

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The production of the L-SBR rubbers is known and is carried out by anionic solution polymerisation using a catalyst based on alkali or alkaline earth metals in an inert organic solvent.

25

The L-SBR rubbers to be used according to the invention usually have molecular weights (numerical averages) of 50,000 to 1,000,000 g/mol and glass transition temperatures of −80 to +20 °C (determined by differential thermoanalysis (DSC)).
As mentioned, the rubber mixtures according to the invention may contain polybutadiene rubbers in addition to the L-SBR rubbers. The polybutadiene rubbers to be used usually have a 1,4-cis content of ≥ 85 %, preferably ≥ 90 % and are preferably produced in the presence of Ziegler/Natta catalysts based on transition metals such as titanium, nickel, cobalt and neodymium. Polybutadiene rubbers of this type are described in: (Houben-Weyl, Methoden der Organischen Chemie, Thieme Verlag, Stuttgart, 1987, Vol. E 20, pages 114 to 134; Ullmann's Encyclopedia of Industrial Chemistry, Vol. A 23, Rubber 3. Synthetic, VCH Verlagsgesellschaft mbH, D-69451 Weinheim, 1993, p. 239-364).

The polybutadiene rubbers to be used according to the invention have molecular weights (numerical averages) of 50,000 to 1,000,000 g/mol and glass transition temperatures of ≤ -100 °C.

Polyisoprene rubber may be used as further rubber component in the rubber mixtures according to the invention, the term polyisoprene rubber covering synthetically produced 1,4-cis polyisoprene (IR) as well as natural rubber (NR). The 1,4-cis content in the polyisoprene rubbers is ≥ 80 %, preferably ≥ 90 %.


The polyisoprene rubbers usually have molecular weights (numerical averages) of 50,000 to 1,000,000 g/mol and a cis-1,4 content > 80 %, preferably > 90 %.

Any known silicas which have also been used and employed hitherto in rubber mixtures may be used as silicas in the rubber mixtures may be used as silicas in the
rubber mixtures according to the invention. Silicas of this type are described, for example, in Ullmann's Encyclopedia of Industrial Chemistry, VCH Verlagsgesellschaft mbH, D-69451 Weinheim, 1993, "Silica", p. 635-645. The silicas may be produced pyrogenically or by precipitation from aqueous solutions, precipitated silicas being preferred. The silicas according to the invention have a specific surface area of about 5 to 1,000 m²/g, preferably of 20 to 400 m²/g. The silicas to be used according to the invention may optionally also exist as mixed oxides with other metal oxides such as aluminium, magnesium, calcium, barium, zinc, zirconium and titanium oxides. The proportion of these metal oxides in the silicas is about 1 to 25 wt.%.

Carbon blacks suitable for the rubber mixtures according to the invention in turn include any carbon blacks known for them, for example those described in Ullmann's Encyclopedia of Industrial Chemistry, VCH Verlagsgesellschaft mbH, D-69451 Weinheim, 1993, Vol. A 5 "Carbon black", p. 95-158. The carbon blacks to be used are produced in the known manner by the gas black, furnace black, lamp black and thermal black processes and are denoted by the new ASTM nomenclature (ASTM D 1765 and D 2516) as: N 110, N 115, N 121, N 125, N 212, N 220, N 231, N 234, N 242, N 293, N 299, S 315, N 326, N 330, N 332, N 339, N 343, N 347, N 351, N 375, N 472, N 539, N 550, N 582, N 630, N 642, N 650, N 660, N 683, N 754, N 762, N 765, N 772, N 774, N 787, N 907, N 908, N 990, N 991. The carbon blacks usually have a BET surface area of about 5 to 200 m²/g, preferably 50 to 150 m²/g.

The term rubber gels, also sometimes as microgels or gels, covers rubber particles which are produced by crosslinking of rubber latices, as described, for example, in US-A-5395891 (BR-gels), US-A-6127488 (SBR-gels) and DE-A 19701487 (NBR-gels). These rubber gels may obviously be produced by corresponding emulsion polymerisation, i.e. without a subsequent crosslinking stage. Agglomerated rubber gels (EP-A 1 078 953) may also be used.
The rubber gels to be used according to the invention may also be modified by incorporation of functional monomers which possess, for example, hydroxyl, carboxyl, vinylpyridine, amino, dialkylamino, chloromethylphenyl and epoxy groups. Rubber gels of this type are described, for example, in DE-A 19 919 459, DE-A-19 834 803 and DE-A-19 834 802.

The rubber gels to be used according to the invention have particle diameters of about 5 to 1,000 nm, preferably 20 to 600 nm (DVN value to DIN 53206). Owing to their crosslinking, they are insoluble and capable of swelling in suitable swelling agents such as toluene. The swelling indices of the rubber gels (Q_i) in toluene are 1 to 15, preferably 1 to 10. The swelling index is calculated from the weight of the solvent-containing gel (after centrifugation at 20,000 rpm) and the weight of the dried gel:

\[ Q_i = \frac{\text{wet weight of the gel}}{\text{dry weight of the gel}}. \]

To determine the swelling index, 250 mg of the gel are allowed to swell for 24 hours in 25 ml toluene while shaking. The gel is then centrifuged off, weighed and then dried at 70 °C to constancy of weight. The dried gel is weighed again. The swelling index is then calculated from the wet weight of the gel and the dry weight of the gel. Rubber gels based on chloroprene rubber, natural rubber, polybutadiene rubber, nitrile rubber and styrene/butadiene rubber are typically used as rubber gels. As already mentioned, the rubber gels may be modified by incorporation of corresponding functional monomers. When modified microgels are used, it is preferable to use those which contain hydroxyl groups.

As already mentioned hereinbefore, some of the normally used silica is replaced by the above-mentioned rubber gels in the rubber mixtures according to the invention. The preferred physical properties of the vulcanisates and the improved processability of the vulcanised rubber mixtures are thus obtained. The aforementioned quantities of rubber gels to be used have been calculated in a first approximation in such a way that the degree of filling of the rubber vulcanisates is
kept virtually constant in terms of volume. On the basis of this calculation with a conventional silica density of 2 g/cm\(^3\) and a rubber gel density of 1 g/cm\(^3\), a substitution of 1 to 20 parts by weight of the silica by 0.5 to 10 parts by weight of rubber gel is obtained.

Filler activators which may be used include sulphur-containing organosilicon compounds having the following basic structures:

\[
\begin{align*}
&\text{R}^1 \\
&\text{R}^2 \text{Si} - \text{Q} - \text{Sx} - \text{R}^4 \\
&\text{R}^3 \\
\end{align*}
\]

\[
\begin{align*}
&\text{R}^1 \\
&\text{R}^2 \text{Si} - \text{Q} - \text{Sx} - \text{Q} - \text{Si} - \text{R}^2 \\
&\text{R}^3 \\
\end{align*}
\]

10

wherein

\( R^1, R^2 \) and \( R^3 \) represent alkoxy groups containing 1 to 10 carbon atoms,

15 \( Q \) represents a spacer group based on aliphatic, heteroaliphatic, aromatic or heteroaromatic carbon chains containing 1 to 20 carbon atoms, preferably 1 to 10 carbon atoms and 1 to 3 heteroatoms such as oxygen, sulphur, nitrogen,

\( R^4 \) represents a radical with the following structures:
wherein $R^5$ represents an aliphatic, heteroaliphatic, cycloaliphatic, aromatic or heteroaromatic radical containing 1 to 20, preferably 1 to 10, carbon atoms and 1 to 3 heteroatoms such as oxygen, nitrogen or sulphur.

Preferred sulphur-containing organosilicon compounds include bis(tri-ethoxy-silylpropyl-polysulphanes) of the following structural formula:

$$\begin{align*}
\text{EtO-Si-CH}_2\text{-CH}_2\text{-CH}_2\left[\text{S}\right]_n\text{-CH}_2\text{-CH}_2\text{-CH}_2\text{-Si-} \text{OEt} \\
\text{OEt} & \quad \text{OEt}
\end{align*}$$

wherein $n = 2$ to 4. Products of this type are commercially available from Degussa under the name Silan Si 75 ($n = 2$) and as Silan Si 69 ($n = 4$).
In addition, the rubber mixtures according to the invention can also contain further rubber auxiliaries and additives which comprise:


The rubber mixtures according to the invention can additionally contain vulcanisation accelerators and crosslinking agents such as sulphur or sulphur donors. Examples of suitable vulcanisation accelerators include mercaptobenzothiazoles, sulphene amides, guanidines, thiurams, dithiocarbamates, thioureas, thiocarbonates and dithiophosphates (cf. Ullmann's Encyclopedia of Industrial Chemistry, VCH Verlagsgesellschaft mbH, D-69451 Weinheim, 1993, Vol. A 23 "Chemicals and Additives", p. 366-417).

The vulcanisation accelerators and crosslinking agents are normally used in quantities of 0.1 to 20 parts by weight, preferably 0.1 to 15 parts by weight, based on the total quantity of rubber, whereas the above-described rubber auxiliaries and additives are used in quantities of 0.5 to 50, preferably 1 to 30 parts by weight, based on the total quantity.

It is obviously possible to mix further components with the rubber mixtures according to the invention such as unmasked and masked, preferably masked bismercaptans of the following general formulae

\[ H-S-Q-S-H, \]

or
\[ X-{\text{Su}}-Q-{\text{Sv}}-Y, \]

wherein

Q represents a spacer group of the aforementioned type and wherein the hydrogen atoms of the unmasked mercaptans may be replaced in a suitable manner by the following substituents X and Y:

\[ \begin{align*}
\text{S} & \quad \text{C} \quad \text{N} \quad R^5 \\
\text{S} & \quad \text{C} \quad \text{S} \quad R^6 \\
\text{O} & \quad \text{C} \quad \text{O} \quad R^5 \\
\text{S} & \quad \text{O} \quad \text{O} \\
\text{N} & \quad \text{C} \quad \text{N} \quad R^1 \\
\end{align*} \]

wherein

\( R^5 \) and Q have the meanings described above,
u and v represents numbers from 1 to 6, preferably u = v = 2.

The following masked bismercaptans are of particular interest:

\[
\text{Na}^+ \quad \begin{array}{c}
\text{O} \\
\text{O}
\end{array} \quad \begin{array}{c}
\text{S} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{CH}_2
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{CH}_2
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{CH}_2
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{CH}_2
\end{array} \quad \begin{array}{c}
\text{S} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{O} \\
\text{O}
\end{array} \quad \text{Na}^+
\]

(A),

and

\[
\left[ \begin{array}{c}
\text{Ph} \\
\text{N}
\end{array} \right] \quad \begin{array}{c}
\text{S} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{S} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{CH}_2
\end{array} \quad \begin{array}{c}
\text{S} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{N} \\
\text{S}
\end{array} \quad \begin{array}{c}
\text{CH}_2 \\
\text{Ph}
\end{array} \quad \left[ \begin{array}{c}
\text{Ph} \\
\text{N}
\end{array} \right]_2
\]

(B),

wherein compound (B) is of particular interest. Compound (B), 1,6-bis-(N,N'-dibenzylthiocarbamoyldithio)-hexane is commercially available under the name Vulcurex®, VP KA 9188 (Bayer AG).

The aforementioned bismercaptans may be used in the mixtures according to the invention in parts by weight of 0.1 to 10, preferably 0.2 to 5, based on 100 parts by weight of component (a).

It is obviously also possible to add further fillers in addition to the aforementioned fillers, silica and carbon black, to the mixtures according to the invention. Examples include:

- synthetic silicates such as aluminium silicate, alkaline-earth silicate such as magnesium silicate or calcium silicate with BET surface areas of 20 to 400 m²/g and primary particle diameters of 5-400 nm,

- natural silicates such as kaolin and other naturally occurring silicas,
- metal oxides such as zinc oxide, calcium oxide, magnesium oxide, aluminium oxide,

- metal carbonates such as calcium carbonate, magnesium carbonate, zinc carbonate,

- metal sulphates such as calcium sulphate, barium sulphate,

- metal hydroxides such as aluminium hydroxide and magnesium hydroxide,

- glass fibres and glass fibre products (laths, strands or glass microspheres),

- thermoplastics (polyamide, polyester, aramide, polycarbonate as well as cellulose and starch).

The aforementioned fillers may be used in quantities of 1 to 20 wt.%, preferably 1 to 10 wt.%, based on the total quantity of silica and carbon black used.

The rubber mixtures according to the invention are produced by mixing the individual components at temperatures in the range of 130 °C to 180 °C, preferably at 145 °C to 180 °C, without the addition of agents capable of initiating vulcanisation of the mixture. Once the components have been mixed at the elevated temperature, agents capable of initiating vulcanisation may be added to the mixture. These include, in particular, crosslinking agents such as sulphur and sulphur-containing compounds as well as vulcanisation accelerators of the aforementioned type. The crosslinking agents are mixed in during the second mixing stage at a temperature ≤ 100 °C, preferably at temperatures in the range of 30 to 60 °C.

The rubber mixtures according to the invention are produced using suitable apparatuses such as mixing rollers, internal mixers or mixing extruders.
Vulcanisation of the rubber mixtures according to the invention is usually carried out at temperatures of about 100 to 250 °C, preferably 130 to 180 °C, optionally under pressure at 10 to 200 bar.

The rubber mixtures produced according to the invention are used for producing tyres and tyre components, in particular for producing tyre treads, subtreads, carcasses, side walls and apex mixtures and also for producing industrial rubber articles such as cushioning elements, roller coverings, conveyor belt coverings, belts, seals, golf ball cores and shoe soles. The mixtures are preferably used for producing tyres and tyre components.
Examples

Production of rubber gels to be used according to the invention

5 The rubber gel (unmodified BR-gel) was produced in the manner described in US-A-5 395 891 (Example BR-gel A1), 2.5 phr (parts per 100 parts of rubber) dicumylperoxide (DCP) being used for crosslinking. The BR-gel A was stabilised and worked up in a similar manner to gel (2) according to DE-A-19 929 347.

10 Gel B was produced in a similar manner to gel (3) in DE-A-19 929 347, 1.5 phr dicumylperoxide (DCP) being used instead of 1.0 phr DCP. Gel B was subjected to hydroxyl modification in a similar manner to gel (2) according to the aforementioned DE-A, 1 phr hydroxyethylmethacrylate (HEMA) rather than 3 phr HEMA being used for hydroxyl modification. Stabilisation and working up were carried out in the above-described manner.

Characteristic data of the starting latices used and the rubber gels produced

<table>
<thead>
<tr>
<th>Product description</th>
<th>Gel type</th>
<th>Styrene content [wt. %]</th>
<th>Quantity of DCP [phr]</th>
<th>Modification</th>
<th>Diameter d50 [nm]</th>
<th>Density [g/cm³]</th>
<th>Gel content [%]</th>
<th>QI</th>
<th>Tg [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Starting latex A</td>
<td>BR</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>111</td>
<td>0.9003</td>
<td>92</td>
<td>15.8</td>
<td>-83</td>
</tr>
<tr>
<td>Gel A</td>
<td>BR</td>
<td>-</td>
<td>2.5</td>
<td>-</td>
<td>110</td>
<td>0.9758</td>
<td>98</td>
<td>3.4</td>
<td>-35</td>
</tr>
<tr>
<td>Starting latex B</td>
<td>SBR</td>
<td>42</td>
<td>-</td>
<td>-</td>
<td>53</td>
<td>0.9673</td>
<td>3.8</td>
<td>26</td>
<td>-32</td>
</tr>
<tr>
<td>Gel B</td>
<td>SBR</td>
<td>42</td>
<td>1.5</td>
<td>-</td>
<td>53</td>
<td>0.9958</td>
<td>91</td>
<td>5.8</td>
<td>-6.5</td>
</tr>
<tr>
<td>Mod. gel B</td>
<td>SBR</td>
<td>42</td>
<td>1.5 HEMA</td>
<td>-</td>
<td>49</td>
<td>1.0031</td>
<td>93</td>
<td>3.3</td>
<td>+4.5</td>
</tr>
</tbody>
</table>

20 The following mixtures are produced on the basis of the above-mentioned gels, and the properties of the corresponding vulcanisates are determined.
Mixture batch A:

The following components are mixed in an internal mixer at 160 °C in the specified sequence (quantities stated in parts by weight)

<table>
<thead>
<tr>
<th>Mixture No.:</th>
<th>1*</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Buna VSL 5025&lt;sup&gt;1)&lt;/sup&gt;</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
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<td>103</td>
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<tr>
<td>Buna CB 24&lt;sup&gt;2)&lt;/sup&gt;</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>Gel A (OBR 1049 B)</td>
<td>0</td>
<td>1</td>
<td>3</td>
<td>5</td>
<td>10</td>
<td>20</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>Renopal 450&lt;sup&gt;3)&lt;/sup&gt;</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>Vulkasil® S&lt;sup&gt;4)&lt;/sup&gt;</td>
<td>80</td>
<td>78</td>
<td>74</td>
<td>70</td>
<td>60</td>
<td>40</td>
<td>20</td>
<td>0</td>
</tr>
<tr>
<td>Silane Si 69®&lt;sup&gt;5)&lt;/sup&gt;</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
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<td>6.4</td>
<td>6.4</td>
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</tr>
<tr>
<td>Stearic acid</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>TMQ&lt;sup&gt;6)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>6PPD&lt;sup&gt;7)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Ozonant wax&lt;sup&gt;8)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

* Comparison

The following components of the compound are mixed into the mixture cooled to 50 °C on the roller:

<table>
<thead>
<tr>
<th>Component</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
<th>1.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zinc oxide</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td>Sulphur</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>CBS&lt;sup&gt;9)&lt;/sup&gt;</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
</tr>
<tr>
<td>DPG&lt;sup&gt;10)&lt;/sup&gt;</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
</tr>
</tbody>
</table>

<sup>1)</sup> Oil-extended solution styrene/butadiene rubber from Bayer AG with 37.5 phr petroleum based on 100 phr solid rubber (vinyl content: 50 wt.%; styrene content: 25 wt.%)

<sup>2)</sup> Neodymium polybutadiene (oil-free) from Bayer AG
3) Plasticiser from Fuchs Mineralölwerke
4) Precipitated active silica from Bayer AG
5) Bis(tri-ethoxy-silylpropyl) tetrasulfan (Si 69® from Degussa AG)
6) 2,2,4-trimethyl-1,2-dihydroquinoline (polym.) Vulkanol® HS from Bayer AG
7) N-1,3-dimethylbutyl-N'-phenyl-p-phenylene-diamine  (Vulkanol® 4020 from Bayer AG
8) Mixture of paraffins and microwaxes (Antilux® 654 from Rheinchemie)
9) Cyclohexylbenzthiazylsulpheneamide (Vulkacit® CZ from Bayer AG)
10) Diphenylguanidine (Vulkacit® D from Bayer AG)

The Mooney viscosity ML 1+4 (100 °C) and the Mooney relaxation (MR 30) are used to characterise the properties of the uncrosslinked compound:

<table>
<thead>
<tr>
<th>Mixture No.:</th>
<th>1*</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>ML 1 + 4 (100 °C) [ME]</td>
<td>111.3</td>
<td>109.7</td>
<td>106.7</td>
<td>98.3</td>
<td>87.5</td>
<td>63.0</td>
<td>48.6</td>
<td>30.4</td>
</tr>
<tr>
<td>MR 30 [%]</td>
<td>24.8</td>
<td>25.4</td>
<td>24.9</td>
<td>22.3</td>
<td>20.0</td>
<td>13.0</td>
<td>9.5</td>
<td>4.3</td>
</tr>
</tbody>
</table>

The vulcanisation behaviour of the mixtures is investigated in a rheometer at 160 °C to DIN 53 529 using the Monsanto rheometer MDR 2000E. Characteristic data such as $F_a$, $F_{max}$, $F_{max}-F_a$, $t_{s1}$, $t_{s0}$ and $t_{90}$ were determined in this way.

<table>
<thead>
<tr>
<th>Mixture No.:</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_a$ [dNm]</td>
<td>5.0</td>
<td>4.6</td>
<td>4.7</td>
<td>4.3</td>
<td>3.6</td>
<td>2.6</td>
<td>1.4</td>
<td>0.9</td>
</tr>
<tr>
<td>$F_{max}$ [dNm]</td>
<td>25.8</td>
<td>25.0</td>
<td>23.4</td>
<td>22.0</td>
<td>19.4</td>
<td>15.4</td>
<td>10.4</td>
<td>7.5</td>
</tr>
<tr>
<td>$F_{max}-F_a$ (dNm)</td>
<td>20.8</td>
<td>20.4</td>
<td>18.7</td>
<td>17.7</td>
<td>15.8</td>
<td>12.8</td>
<td>9.0</td>
<td>6.6</td>
</tr>
<tr>
<td>$t_{s1}$ [min.]</td>
<td>0.6</td>
<td>0.4</td>
<td>0.9</td>
<td>1.1</td>
<td>1.4</td>
<td>1.8</td>
<td>2.2</td>
<td>2.4</td>
</tr>
<tr>
<td>$t_{s0}$ [min.]</td>
<td>3.7</td>
<td>3.9</td>
<td>4.0</td>
<td>4.0</td>
<td>3.7</td>
<td>3.2</td>
<td>3.1</td>
<td>3.5</td>
</tr>
<tr>
<td>$t_{90}$ [min.]</td>
<td>10.7</td>
<td>10.4</td>
<td>10.1</td>
<td>9.7</td>
<td>8.7</td>
<td>9.4</td>
<td>9.5</td>
<td>8.0</td>
</tr>
</tbody>
</table>

DIN 53 529, part 3 gives the following meanings:
The mixtures are vulcanised in the press at 165 °C for 20 minutes.

The following test values are found on the vulcanisates:

<table>
<thead>
<tr>
<th>Mixture No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile strength [MPa]</td>
<td>17.6</td>
<td>18.4</td>
<td>16.1</td>
<td>17.0</td>
<td>13.7</td>
<td>11.5</td>
<td>8.9</td>
<td>7.0</td>
</tr>
<tr>
<td>Elongation at break [%]</td>
<td>373</td>
<td>370</td>
<td>355</td>
<td>375</td>
<td>345</td>
<td>380</td>
<td>475</td>
<td>600</td>
</tr>
<tr>
<td>Modulus/50 % elongation [MPa]</td>
<td>1.8</td>
<td>1.8</td>
<td>1.6</td>
<td>1.6</td>
<td>1.4</td>
<td>0.9</td>
<td>0.7</td>
<td>0.6</td>
</tr>
<tr>
<td>Modulus/100 % elongation [MPa]</td>
<td>3.4</td>
<td>3.6</td>
<td>3.2</td>
<td>3.2</td>
<td>2.9</td>
<td>2.2</td>
<td>1.1</td>
<td>1.0</td>
</tr>
<tr>
<td>Modulus/300 % elongation [MPa]</td>
<td>13.2</td>
<td>13.8</td>
<td>12.8</td>
<td>12.5</td>
<td>11.3</td>
<td>8.0</td>
<td>4.2</td>
<td>2.5</td>
</tr>
<tr>
<td>Shore A hardness, 23 °C</td>
<td>69</td>
<td>67</td>
<td>65</td>
<td>64</td>
<td>62</td>
<td>57</td>
<td>47</td>
<td>40</td>
</tr>
<tr>
<td>Rebound elasticity 23 °C [%]</td>
<td>28</td>
<td>30</td>
<td>31</td>
<td>32</td>
<td>37</td>
<td>44</td>
<td>49</td>
<td>39</td>
</tr>
<tr>
<td>Rebound elasticity 70 °C [%]</td>
<td>57</td>
<td>59</td>
<td>61</td>
<td>62</td>
<td>66</td>
<td>67</td>
<td>68</td>
<td>65</td>
</tr>
<tr>
<td>DIN abrasion [mm²]</td>
<td>90</td>
<td>90</td>
<td>90</td>
<td>95</td>
<td>105</td>
<td>150</td>
<td>220</td>
<td>335</td>
</tr>
<tr>
<td>tan δ (0 °C)</td>
<td>0.486</td>
<td>0.514</td>
<td>0.548</td>
<td>0.534</td>
<td>0.536</td>
<td>0.501</td>
<td>0.290</td>
<td>0.356</td>
</tr>
<tr>
<td>tan δ (23 °C)</td>
<td>0.240</td>
<td>0.226</td>
<td>0.213</td>
<td>0.203</td>
<td>0.171</td>
<td>0.146</td>
<td>0.130</td>
<td>0.192</td>
</tr>
<tr>
<td>tan δ (60 °C)</td>
<td>0.102</td>
<td>0.096</td>
<td>0.090</td>
<td>0.087</td>
<td>0.072</td>
<td>0.060</td>
<td>0.065</td>
<td>0.088</td>
</tr>
</tbody>
</table>

Result: Mixture batch A shows that by substituting 1, 3 and 5 phr of unmodified BR-gel for 2, 6 and 10 phr of silica respectively, significant advantages may be achieved in the wet slip behaviour (tan δ/0 °C) and in the rolling resistance (tan δ/60 °C)
without significant losses in processability (compound viscosity) and in abrasion resistance.

**Mixture batch B:**

5

The following components are mixed in an internal mixer at 160 °C in the specified sequence (quantities stated in parts by weight)

<table>
<thead>
<tr>
<th>Mixture No.</th>
<th>1*</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Buna VSL 5025&lt;sup&gt;1)&lt;/sup&gt;</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
<td>103</td>
</tr>
<tr>
<td>Buna CB 24&lt;sup&gt;2)&lt;/sup&gt;</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>Gel B (OBR 1046 A)</td>
<td>0</td>
<td>1</td>
<td>3</td>
<td>5</td>
<td>10</td>
<td>20</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>Renopal 450&lt;sup&gt;3)&lt;/sup&gt;</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>Vulkasil® S&lt;sup&gt;4)&lt;/sup&gt;</td>
<td>80</td>
<td>78</td>
<td>74</td>
<td>70</td>
<td>60</td>
<td>40</td>
<td>20</td>
<td>0</td>
</tr>
<tr>
<td>Silane Si 69&lt;sup&gt;5)&lt;/sup&gt;</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
<td>6.4</td>
</tr>
<tr>
<td>Stearic acid</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>TMQ&lt;sup&gt;6)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>6PPD&lt;sup&gt;7)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Ozonant wax&lt;sup&gt;8)&lt;/sup&gt;</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

* Comparison

The following components of the compound are mixed into the mixture cooled to 50 °C on the roller:

<table>
<thead>
<tr>
<th></th>
<th>2.5</th>
<th>2.5</th>
<th>2.5</th>
<th>2.5</th>
<th>2.5</th>
<th>2.5</th>
<th>2.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zinc oxide</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td>Sulphur</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
<td>1.5</td>
</tr>
<tr>
<td>CBS&lt;sup&gt;9)&lt;/sup&gt;</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
<td>1.7</td>
</tr>
<tr>
<td>DPG&lt;sup&gt;10)&lt;/sup&gt;</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
</tr>
</tbody>
</table>

<sup>1)</sup> Solution styrene/butadiene rubber from Bayer AG with 37.5 phr petroleum (vinyl content: 50 wt.%; styrene content: 25 wt.%)
2) Neodymium polybutadiene (oil-free) from Bayer AG
3) Plasticiser from Fuchs Mineralölwerke
4) Precipitated active silica from Bayer AG
5) Bis(tri-ethoxy-silylpropyl) tetrasulfan (Si 69® from Degussa AG)
6) 2,2,4-trimethyl-1,2-dihydroquinoline (polym.) Vulkanox® HS from Bayer AG
7) N-1,3-dimethylbutyl-N'-phenyl-p-phenylene-diamine (Vulkanox® 4020 from Bayer AG
8) Mixture of paraffins and microwaxes (Antilux® 654 from Rheinchemie)
9) Cyclohexylbenzthiazysulspheneamide (Vulkacit® CZ from Bayer AG)
10) Diphenylguanidine (Vulkacit® D from Bayer AG)

The Mooney viscosity ML 1+4 (100 °C) and the Mooney relaxation (MR 30) are used to characterise the properties of the uncrosslinked compound:

<table>
<thead>
<tr>
<th>Mixture No.</th>
<th>1*</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>ML 1 + 4 (100 °C) [ME]</td>
<td>111.3</td>
<td>107.5</td>
<td>104.8</td>
<td>89.0</td>
<td>82.7</td>
<td>60.4</td>
<td>46.7</td>
<td>31.7</td>
</tr>
<tr>
<td>MR 30 [%]</td>
<td>24.8</td>
<td>24.0</td>
<td>24.2</td>
<td>24.7</td>
<td>18.9</td>
<td>12.6</td>
<td>9.6</td>
<td>4.1</td>
</tr>
</tbody>
</table>

The vulcanisation behaviour of the mixtures is investigated in a rheometer at 160 °C to DIN 53 529 using the Monsanto rheometer MDR 2000E. Characteristic data such as $F_a$, $F_{max}$, $F_{max}-F_a$, $t_{s1}$, $t_{50}$ and $t_{90}$ were determined in this way.
<table>
<thead>
<tr>
<th>Mixture No.:</th>
<th>1st</th>
<th>2nd</th>
<th>3rd</th>
<th>4th</th>
<th>5th</th>
<th>6th</th>
<th>7th</th>
<th>8th</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_a$ [dNm]</td>
<td>5.0</td>
<td>5.1</td>
<td>4.6</td>
<td>4.3</td>
<td>3.3</td>
<td>2.1</td>
<td>1.4</td>
<td>0.9</td>
</tr>
<tr>
<td>$F_{max}$ [dNm]</td>
<td>25.8</td>
<td>24.2</td>
<td>22.2</td>
<td>20.7</td>
<td>17.9</td>
<td>14.5</td>
<td>9.6</td>
<td>6.7</td>
</tr>
<tr>
<td>$F_{max}-F_a$ [dNm]</td>
<td>20.8</td>
<td>19.1</td>
<td>17.6</td>
<td>16.4</td>
<td>14.6</td>
<td>12.4</td>
<td>8.2</td>
<td>5.8</td>
</tr>
<tr>
<td>$t_{s1}$ [min.]</td>
<td>0.6</td>
<td>0.9</td>
<td>0.9</td>
<td>1.3</td>
<td>1.5</td>
<td>1.9</td>
<td>2.2</td>
<td>2.3</td>
</tr>
<tr>
<td>$t_{50}$ [min.]</td>
<td>3.7</td>
<td>4.1</td>
<td>4.0</td>
<td>4.1</td>
<td>3.8</td>
<td>3.3</td>
<td>3.0</td>
<td>3.1</td>
</tr>
<tr>
<td>$t_{90}$ [min.]</td>
<td>10.7</td>
<td>10.5</td>
<td>10.1</td>
<td>9.0</td>
<td>8.0</td>
<td>8.7</td>
<td>9.2</td>
<td>9.1</td>
</tr>
</tbody>
</table>

DIN 53 329, part 3 gives the following meanings:

5 $F_a$: vulcameter display in the minimum of crosslinking isotherms

$F_{max}$: maximum of vulcameter display

$F_{max}-F_a$: difference in vulcameter displays between maximum and minimum

$t_{s1}$: time when the vulcameter display has increased by 1 Nm relative to the minimum

10 $t_{50}$: time when 50% of conversion are achieved

$t_{90}$: time when 90% of conversion are achieved

The mixtures are vulcanised in the press at 165 °C for 20 minutes.
The following test values are found on the vulcanisates:

<table>
<thead>
<tr>
<th>Mixture No.:</th>
<th>1*</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile strength [MPa]</td>
<td>17.6</td>
<td>17.7</td>
<td>17.5</td>
<td>15.3</td>
<td>14.2</td>
<td>10.1</td>
<td>6.6</td>
<td>5.1</td>
</tr>
<tr>
<td>Elongation at break [%]</td>
<td>373</td>
<td>375</td>
<td>380</td>
<td>345</td>
<td>370</td>
<td>370</td>
<td>435</td>
<td>540</td>
</tr>
<tr>
<td>Modulus/50 % elongation [MPa]</td>
<td>1.8</td>
<td>1.8</td>
<td>1.6</td>
<td>1.6</td>
<td>1.4</td>
<td>1.1</td>
<td>0.7</td>
<td>0.6</td>
</tr>
<tr>
<td>Modulus/100 % elongation [MPa]</td>
<td>3.4</td>
<td>3.4</td>
<td>3.2</td>
<td>3.1</td>
<td>2.7</td>
<td>2.0</td>
<td>1.1</td>
<td>0.9</td>
</tr>
<tr>
<td>Modulus/300 % elongation [MPa]</td>
<td>13.2</td>
<td>13.0</td>
<td>12.7</td>
<td>12.4</td>
<td>10.3</td>
<td>7.3</td>
<td>3.5</td>
<td>2.1</td>
</tr>
<tr>
<td>Shore A hardness, 23 °C</td>
<td>69</td>
<td>67</td>
<td>66</td>
<td>64</td>
<td>60</td>
<td>55</td>
<td>46</td>
<td>40</td>
</tr>
<tr>
<td>Rebound elasticity 23 °C [%]</td>
<td>28</td>
<td>29</td>
<td>29</td>
<td>30</td>
<td>32</td>
<td>33</td>
<td>43</td>
<td>34</td>
</tr>
<tr>
<td>Rebound elasticity 70 °C [%]</td>
<td>57</td>
<td>59</td>
<td>60</td>
<td>63</td>
<td>65</td>
<td>69</td>
<td>72</td>
<td>72</td>
</tr>
<tr>
<td>DIN abrasion [mm³]</td>
<td>90</td>
<td>80</td>
<td>70</td>
<td>80</td>
<td>90</td>
<td>130</td>
<td>225</td>
<td>-</td>
</tr>
<tr>
<td>tan δ (0 °C)</td>
<td>0.486</td>
<td>0.488</td>
<td>0.511</td>
<td>0.514</td>
<td>0.526</td>
<td>0.480</td>
<td>0.312</td>
<td>0.358</td>
</tr>
<tr>
<td>tan δ (23 °C)</td>
<td>0.240</td>
<td>0.229</td>
<td>0.222</td>
<td>0.212</td>
<td>0.201</td>
<td>0.211</td>
<td>0.191</td>
<td>0.248</td>
</tr>
<tr>
<td>tan δ (60 °C)</td>
<td>0.102</td>
<td>0.098</td>
<td>0.089</td>
<td>0.082</td>
<td>0.070</td>
<td>0.057</td>
<td>0.065</td>
<td>0.070</td>
</tr>
</tbody>
</table>

Result: Mixture batch B shows that by substituting 1, 3 and 5 phr of unmodified SBR-gel for 2, 6 and 10 phr of silica respectively, significant advantages may be achieved in the wet slip behaviour (tan δ/0 °C) and in the rolling resistance (tan δ/60 °C) without significant losses in processability (compound viscosity) and in abrasion resistance.
Claims

1. Rubber mixtures consisting of

   a) solution styrene/butadiene rubber (L-SBR)

   b) silica

   c) carbon black and

   d) rubber gel,

wherein component a) is present in the mixture in a quantity of 100 parts by weight, component b) in quantities of 20 to 120 parts by weight, component c) in quantities of 5 to 60 parts by weight and component d) in quantities of 0.1 to 10 parts by weight, providing that the proportion of component c) is 50 wt.% maximum based on the proportion of component b).

2. Rubber mixtures according to claim 1, characterised in that, in addition to L-SBR rubbers, they contain polybutadiene rubber (BR) and polyisoprene rubber (IR and NR) both individually and in combination with one another, the proportion of the polybutadiene rubber, the proportion of the polyisoprene rubber or the proportion of the combination of polybutadiene rubber and polyisoprene rubber in the mixture is 1 to 50 wt.%, providing that 5 to 95 wt.% of polybutadiene rubber based on the total combination of polybutadiene rubber and polyisoprene rubber are present when using the combination of polybutadiene rubber and polyisoprene rubber.

3. Rubber mixtures according to claim 1, characterised in that they contain a filler activator based on organosilicon sulphur compounds (component e)) in quantities of 0.1 to 15 parts by weight.

4. Rubber mixtures according to claim 1, characterised in that they contain the known and conventional rubber auxiliaries and additives as well as cross-linking
agents and vulcanisation accelerators (component g)) in quantities of 0.05 to 80 parts by weight.

5. Process for producing the rubber mixtures according to claim 1, characterised in that the mixture components are blended at temperatures in the range of 130 to 180 °C without the addition of vulcanisation-initiating agents and the agents of the mixture which initiate vulcanisation of the mixture are then added at temperatures $\leq 100$ °C.

6. Use of the rubber mixtures according to claim 1 for producing tyres and tyre components and for producing industrial rubber articles.