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(54) Titre : MOULURES BASEES SUR DES PRODUITS DE REACTION ENTRE LES POLYOLS ET LES ISOCYANATES
(54) Title: MOULDINGS BASED ON REACTION PRODUCTS OF POLYOLS AND ISOCYANATES

(57) **Abrégé/Abstract:**

The present invention relates to moulding compositions comprising the following components: a) one or more polyisocyanates, b) one or more polyols, c) one or more catalysts catalysing the reaction between polyisocyanate and polyol and d) one or more pyrogenically produced oxides of a metal or of a metalloid, characterized in that the moulding composition comprises, as further component, e) at least one or more siloxanes, mouldings obtainable with use of the said moulding compositions, and also a process for the production of the mouldings.



Abstract

The present invention relates to moulding compositions comprising the following components: a) one or more polyisocyanates, b) one or more polyols, c) one or more catalysts catalysing the reaction between polyisocyanate and polyol and d) one or more pyrogenically produced oxides of a metal or of a metalloid, characterized in that the moulding composition comprises, as further component, e) at least one or more siloxanes, mouldings obtainable with use of the said moulding compositions, and also a process for the production of the mouldings.

E V O N I K I n d u s t r i e s A G, Essen
T e c h n o g e l G m b H, Duderstadt

Mouldings based on reaction products of polyols and
isocyanates

The present invention relates to moulding compositions comprising the following components: a) one or more polyisocyanates, b) one or more polyols, c) one or more catalysts catalysing the reaction between
5 polyisocyanate and polyol and d) one or more pyrogenically produced oxides of a metal or of a metalloid, characterized in that the moulding composition comprises, as further component, e) at least one or more siloxanes, mouldings obtainable with
10 use of the said moulding compositions, and also a process for the production of the mouldings.

Prior art:

WO 2009/092505 describes the production of insulating
15 polyurethane foams, where porous solids, in particular zeolites, are used as nucleating agents.

EP 0057838 A1 describes the production of gel cushions, where the gel is composed of a high-molecular-weight
20 matrix made of covalently crosslinked polyurethane, and, securely bound within the matrix via secondary valence forces, liquid dispersion medium made of one or more polyhydroxy compounds. The gel cushions are produced by casting the reaction mixture into prepared
25 casings, where the gel reaction is completed.

EP 0511570 A1 describes gels based on reaction products of polyols and polyisocyanates, where polyols used
comprise mixtures of polyols with different hydroxy
30 numbers. Again, the gels are produced by casting the

reaction mixtures into prepared casings, where the gel reaction is concluded.

EP 1125975 A1 describes the production of gels based on
5 reaction products of polyisocyanates and polyols which
comprise at least one pyrogenically produced oxide of a
metal or metalloid in order to improve mechanical
tensile strength and elongation at break. Here again,
the shaping of the gel is achieved by casting the
10 reaction mixture into a casing or mould.

In Rheol. Acta (2005) 44, 644-653 (DOI 10.2007/s00397-
005-0446-3), Saint Michel et al. describe the changes
in the rheological properties of mixtures of a silica-
15 gelled polymer matrix comprising polyol, catalyst and
surfactant caused by addition of CaCO₃ particles with a
particle size in the micrometre range. Surfactant used
comprises a polydimethylsiloxane/polyoxyethylene
copolymer. According to the first complete paragraph in
20 the right-hand column on page 650 of that document, the
surfactant was found to have no effect on rheological
properties.

A disadvantage of the gels described and/or of the
25 process for production thereof is restriction to
mouldings which are obtainable via casting and
optionally subsequent trimming.

It was an object of the present invention to provide a
30 process for production of gels which provides, in a
simple manner, access to a wider variety of shapes.

Surprisingly, it has been found that addition of
organomodified siloxanes to the reaction mixture can
35 render this thixotropic or more thixotropic, and thus
permits moulding of the reaction mixtures in a simple
manner via conventional shaping processes such as

extrusion, to give dimensionally stable linear products.

The present invention therefore provides moulding
5 compositions comprising the following components: a)
one or more polyisocyanates, b) one or more polyols, c)
one or more catalysts catalysing the reaction between
polyisocyanate and polyol and d) one or more
10 pyrogenically produced oxides of a metal or of a
metalloid, where a feature of these is that the
moulding composition comprises, as further component,
e) at least one or more siloxanes of the formula (I),
mouldings obtainable with use of the said moulding
compositions, and also a process for the production of
15 the mouldings.

An advantage of the claimed moulding compositions is
that they can be moulded in a simple manner by using
conventional shaping processes to give dimensionally
20 stable mouldings. The said dimensionally stable
mouldings then react to give the desired final
products.

The claimed moulding compositions can in particular
25 have the advantage of a higher yield point than
moulding compositions of this type which comprise no
compound of the formula (I).

If the moulding compositions comprise no blowing agents
30 and are intended for processing to give gel-type
mouldings, the claimed moulding compositions have the
further advantage that the mouldings obtained after the
gel reaction correspond to, or at least in essence
correspond to, the dimensionally stable mouldings
35 previously obtained from the shaping process, i.e. that
the change in all of the dimensions is smaller than
20%. It is therefore possible where appropriate, or

even frequently, to avoid a downstream mechanical operation, in particular subsequent shaping.

An advantage of the claimed process is that by virtue
5 of controlled addition of the siloxane it is possible
to achieve controlled adjustment of the rheological
properties of the system. It is thus possible when the
product discharged is linear to modify the
concentration of the siloxane of the formula (I)
10 appropriately in such a way as to permit production of
a smooth transition between the start and end of a
(circular) sealing bead.

Another advantage of the claimed process is that it is
15 readily possible to achieve controlled adjustment of
thixotropy/viscosity (switch-on and switch-off or
controlled adjustment of thixotropy or viscosity). This
can be achieved via simple addition of a liquid
component (siloxane of the formula (I)), and this also
20 results in simple processing. The conventional methods
are very inconvenient, and conclusive, i.e. produce a
fixed viscosity.

The subject matter of the invention is described by way
25 of example below, without any intention that the
invention be restricted to these examples of
embodiments. Insofar as ranges, general formulae or
classes of compound are stated below, these are
intended to encompass not only the appropriate ranges
30 or groups of compounds explicitly mentioned but also
all of the sub-ranges and sub-groups of compounds which
can be obtained via extraction of individual values
(ranges) or compounds. Where documents are cited for
the purposes of the present description, the entire
35 content of these is intended to become part of the
disclosure of the present invention, in particular in
relation to the factual context in connection with

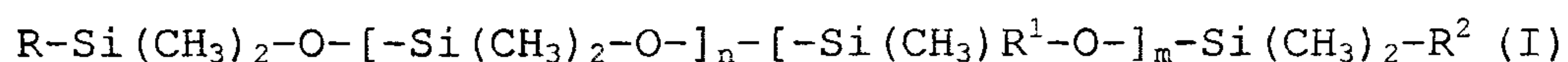
which the document has been cited. Percentage data are in percent by weight unless otherwise stated. Where average values are stated below, they are weight averages unless otherwise stated. Where parameters are stated below that have been determined by measurement, the measurements were made at a temperature of 25°C and at a pressure of 101 325 Pa unless otherwise stated.

For the purposes of the present invention, polyurethane (PU) is a product obtainable via reaction of isocyanates and polyols or compounds having groups reactive towards isocyanate. It is also possible here that other functional groups are formed alongside the polyurethane that gives the material its name, examples being allophanates, biurets, ureas, and isocyanurates. For the purposes of the present invention, PU therefore means not only polyurethane but also polyisocyanurate.

The claimed moulding compositions, comprising the following components:

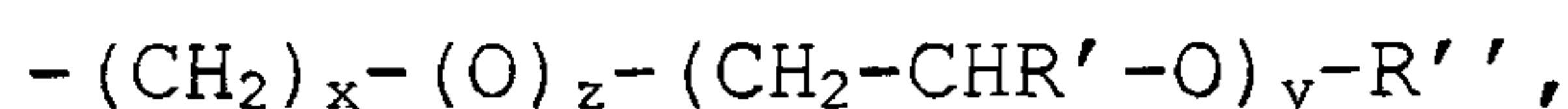
- a) one or more polyisocyanates,
 - b) one or more polyols,
 - c) one or more catalysts catalysing the reaction between polyisocyanate and polyol and
 - d) one or more pyrogenically produced oxides of a metal or of a metalloid,
- are characterized in that the moulding compositions comprise, as further component,
- e) at least one or more siloxanes of the formula (I)

30



where

- R, R¹ and R² are identical or different,
- R and/or R² are methyl or R¹,
- R¹, being identical or different, is selected from the group of



$-(\text{CH}_2)-\text{R}''''$,
 $-(\text{CH}_2)_x-(\text{O})_z-\text{R}''$,
 $-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{O}-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2\text{OH}$
 $-(\text{CH}_2)_x-(\text{O})_z-(\text{CH}_2-\text{CHR}'-\text{O})_y-\text{R}''$,

5 R' , being identical or different, is $-\text{H}$, $-\text{CH}_3$, $-\text{CH}_2\text{CH}_3$, or phenyl moiety,

R'' , being identical or different, is $-\text{H}$, $-(\text{CO})-\text{R}''''$,
 $-(\text{CO})-\text{NH}-\text{R}''''$ or $-\text{alkyl}$, preferably C_1 to C_{40} -alkyl,
 with preference C_1 - or C_6 to C_{30} -alkyl,

10 R'''' , being identical or different, is C_1 to C_{40} -alkyl, $-\text{aryl}$ or $-\text{alkylaryl}$; optionally also substituted with halogens,

$n+m+2 =$ from 10 to 150, preferably from 12 to 85, with particular preference from 15 to 47,

15 $m =$ from 0 to 20, preferably from 1 to 4,

$x =$ from 2 to 15, preferably from 3 to 10,

$y =$ from 1 to 40, preferably from 2 to 19,

$z =$ 0 or 1,

20 where the units $(\text{CH}_2-\text{CHR}'-\text{O})$ can be identical or different,

with the proviso that for $m = 0$ at least one moiety R or R^2 is identical with R^1 ,

and that if $z=0$ x and $y = 0$ and R'' has at least 2 carbon atoms,

25 with the proviso that the moulding composition comprises less than 2% by weight, preferably less than 0.2% by weight, based on the entire constitution of the moulding composition, of physical blowing agent.

30 It is preferable that the claimed moulding compositions comprise, as component e), siloxane compounds of the formula (I) in which the moieties R^1 are $-(\text{CH}_2)_x-(\text{O})_z-(\text{CH}_2-\text{CHR}'-\text{O})_y-\text{R}''$ moieties, where R' , R'' , x , y , and z are as defined above. It is particularly preferable

35 that the numeric-average molar proportion of alkylene oxide units where $\text{R}' = \text{H}$, based on all of the alkylene oxide units in the moieties R^1 , is at least 80%,

preferably at least 90%, and/or that at least 80% of the moieties R'' are hydrogen.

Very particularly preferred claimed moulding compositions comprise, as component e), siloxane compounds of the formula (I) in which the quotient $Q = \text{number of Si atoms/number of moieties } R^1$ (in each case in numeric average per molecule) is from more than 5 to less than or equal to 16, preferably from more than 8 to less than or equal to 12.

The proportion of component d) in the claimed moulding composition is preferably from 1 to 30 parts by weight, preferably from 5 to 20 parts by weight, per 100 parts by weight of component b) and/or, with preference and, the proportion of component e) in the moulding composition is preferably from 0.05 to 10 parts by weight, preferably from 0.2 to 5 parts by weight, with particular preference from 0.5 to 3 parts by weight, per 100 parts by weight of component b).

Component a)

In principle, it is possible to use any of the aliphatic, cycloaliphatic and aromatic isocyanates, and their modified forms and prepolymers. The polyisocyanates used can comprise one or more isocyanate groups, but it is preferable that the isocyanates should be at least difunctional. Particularly suitable isocyanates for the production of moulding compositions, in particular of gels, are described by way of example in EP 1 125 975 A1. Examples of isocyanates very particularly suitable for the production of gels are toluene 2,4-diisocyanate (TDI), methylenediphenyl diisocyanate (MDI), hexamethylene diisocyanate (HMDI), isophorone diisocyanate (IPDI) and 4,4'-diisocyanatodicyclohexylmethane (H12MDI). It is particularly preferable to use prepolymeric isocyanates

based on HDI and MDI with functionalities of from 2 to 4.

Component b)

5 It is possible to use, as component b), any of the compounds that are reactive toward isocyanate groups, or a blend of these compounds. It is preferable to use, as component b) for the production of the claimed moulding compositions, in particular gels, the polyols
10 described in EP 1125975 A1. It is preferable to use, as components b), polyether polyols with OH number < 100 and with a functionality of from 2 to 8, particularly those with OH number < 60 and with a functionality of from 3 to 6.

15

Component c)

It is possible to use, as component c), any of the catalysts which catalyse the reaction between isocyanate groups and groups reactive toward
20 isocyanate.

Examples of catalysts that can be used are tertiary amines, such as triethylamine, tributylamine, N-methylmorpholine, N-ethylmorpholine, N,N,N,N-teramethyl-
25 ethylenediamine, 1,4-diazabicyclo[2.2.2]octane, N-methyl-N-dimethylaminoethylpiperazine, N,N-dimethylbenzylamine, bis(N,N-diethylaminoethyl) adipate, pentamethyldiethylenetriamine, N,N-dimethylcyclohexylamine, N,N,N',N'-tetramethyl-1,3-butanediamine, N,N-
30 dimethyl-β-phenylethylamine, 1,2-dimethylimidazole and 2-methylimidazole, Mannich bases derived from secondary amines, e.g. dimethylamine, and from aldehydes, preferably formaldehyde, or from ketones, such as acetone, methyl ethyl ketone or cyclohexanone, and from
35 phenols, such as phenol or nonyl phenol, or from bisphenols; other examples are sila-amines having carbon-silicon bonds as described by way of example in DE-C 1 229 290 and US Patent 3 620 984, preferably

2,2,4-trimethyl-2-silamorpholine or 1,3-diethylamino-
methyltetramethyldisiloxane, nitrogen-containing bases,
preferably tetralkylammonium hydroxides, alkali metal
hydroxides, preferably sodium hydroxide, alkali metal
5 phenolates, preferably sodium phenolate, and alkali
metal alcoholates, preferably sodium methoxide
(methylate), hexahydrothiazines, and organometallic
compounds, in particular organic compounds of, and
organic salts of, tin, zirconium, bismuth, titanium and
10 zinc.

Other catalysts, and also details of the mode of action
of the catalysts, are described in Kunststoff-Handbuch
[Plastics handbook] volume VII, edited by Vieweg and
Höchtlen, Carl-Hanser-Verlag Munich, 1966, e.g. on pp.
15 96-102. It is also possible to use mixtures of various
catalysts.

An example of an amount that can be used of the
catalyst is from 0.01 to 10% by weight, based on the
total weight of the mixture for production of the gel.

20

Component c) present in the claimed moulding
composition preferably comprises tertiary amines, in
particular one of the abovementioned amines, and/or
organic tin salts, organic bismuth salts or organic
25 zinc salts, in particular of carboxylic acids having
from 8 to 18 carbon atoms. Component c) present in the
claimed moulding composition (for the production of
gels) particularly preferably comprises bismuth tris-
neodecanoate.

30

Component d)

The pyrogenically produced oxides can preferably be
produced by the flame hydrolysis route. Pyrogenically
produced oxides of metals and/or of metalloids are
35 known from Ullmanns Enzyklopädie der technischen Chemie
[Ullmann's Encyclopaedia of industrial chemistry] 4th
Edition, volume 21, pp. 464 and 465 (1982). The claimed

moulding compositions can comprise physical mixtures of pyrogenically produced oxides of metals and/or of metalloids and/or can comprise pyrogenically produced mixed oxides of metals and/or of metalloids.

5

It is preferable to use one or more of the following pyrogenically produced oxides: silicon dioxide, aluminium oxide, mixed oxide of silicon dioxide and aluminium oxide, titanium dioxide, mixed oxide of titanium dioxide and iron oxide. Component d) present in the claimed moulding composition preferably comprises silicon dioxide.

Particular preference is given to use, as component d), of pyrogenically produced silicon dioxides which have been surface-treated, examples being Aerosil[®] R 8200, Aerosil[®] R972, Aerosil[®] R974, Aerosil[®] R805, Aerosil[®] R 202 or Aerosil[®] R812. These oxides are described in the series of publications entitled Pigmente [Pigments] No. 27 (August 1993) from Degussa AG.

20

Examples of surface-treating agents that can be used for the pyrogenically produced oxides are: dimethyldichlorosilane, trimethyldichlorosilane, hexamethyldisilazane, polydimethylsiloxanes, alkylsilanes, for example trimethoxyoctylsilane and triethoxyoctylsilane.

In particular, the pyrogenically produced hydrophilic and, respectively, hydrophobic oxides stated in Tables 1a and 1b can be used:

30

Table 1a: Hydrophilic oxides

| Test method | AEROSIL 90 | AEROSIL 130 | AEROSIL 150 | AEROSIL 200 | AEROSIL 300 | AEROSIL 380 | AEROSIL OX 50 | AEROSIL TT 600 | AEROSIL MOX 80 | AEROSIL MOX 170 | AEROSIL COK 84 | |
|---|---------------------------|-------------|-------------|--------------------|-------------|-------------|---------------|----------------|----------------|-----------------|----------------|--------|
| Behaviour with respect to water | hydrophilic | | | | | | | | | | | |
| Appearance | free-flowing white powder | | | | | | | | | | | |
| BET¹⁾ surface area | m ² /g | 90+15 | 130+25 | 150+15 | 200+25 | 300+30 | 380+30 | 50+15 | 200+50 | 80+20 | 170+30 | 170+30 |
| Average size of primary particles | nm | 20 | 16 | 14 | 12 | 7 | 7 | 40 | 40 | 30 | 15 | - |
| Tamped density (approx. value)²⁾ | g/l | 80 | 50 | 50 | 50 | 50 | 50 | 130 | 60 | 60 | 50 | 50 |
| compacted product (suffix "V")¹¹⁾ | g/l | 120 | 120 | 120 | 120 | 120 | 130 | | | | | |
| VV product (suffix "VV")¹²⁾ | g/l | | | 50/75 | 50/75 | 50/75 | | | | | | |
| Loss on drying³⁾ (2 hours at 105°C) on leaving supplier's works | % | <1.0 | <1.5 | <0.5 ⁹⁾ | <1.5 | <1.5 | <2.0 | <1.5 | <2.5 | <1.5 | <1.5 | <1.5 |
| Loss on ignition^{4) 7)} (2 hours at 1000°C) | % | <1 | <1 | <1 | <1 | <2 | <2.5 | <1 | <2.5 | <1 | <1 | <1 |
| pH⁵⁾ | | 3.7- | 3.7- | 3.7- | 3.7-4.7 | 3.7-4.7 | 3.7- | 3.8- | 3.6- | 3.6- | 3.6- | 3.6- |
| | | 4.7 | 4.7 | 4.7 | | 4.7 | 4.7 | 4.8 | 4.5 | 4.5 | 4.5 | 4.3 |

| | | | | | | | | | | | | |
|---|---|--------|--------|--------|--------|--------|--------|--------|--------|-------------|-------------|-------|
| SiO₂ ⁸⁾ | % | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >98.3 | >98.3 | 82-86 |
| AlO₂O₃ ⁸⁾ | % | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | 0.3- 1.3 | 0.3- 1.3 | 14-18 |
| Fe₂O₃ ⁸⁾ | % | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.01 | <0.1 |
| TiO₂ ⁸⁾ | % | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 |
| HCl ^{8) 10)} | % | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.025 | <0.1 |
| Sieve residue ⁶⁾ | % | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.1 | <0.1 |

Mocker method, 45 mm)

- 1) based on DIN 66131
- 2) based on DIN ISO 787/XI, JIS K 5101/18 (not sieved)
- 3) based on DIN ISO 787/II, ASTM D 280, JIS K 5101/21
- 4) based on DIN 55921, ASTM D 1280, JIS K 5101/23
- 5 5) based on DIN ISO 787/IX, ASTM D 1208, JIS K 5101/24
- 6) based on DIN ISO 787/XVIII, JIS K 5101/20
- 7) based on substance dried at 105°C for 2 hours
- 8) based on substance ignited at 1000°C for 2 hours
- 9) specific packaging providing protection from moisture
- 10 10) HCl content is a constituent of the loss on ignition
- 11) V product is supplied in 20 kg bags
- 12) VV product is currently supplied exclusively from the Rheinfelden works

Table 1b: Hydrophobic oxides

| Test method | AEROSIL R 972 | AEROSIL R 974 | AEROSIL R 202 | AEROSIL R 805 | AEROSIL R 812 | AEROSIL R 812 S | AEROSIL R 104 | AEROSIL R 106 | |
|---|---------------------------|------------------|------------------|------------------|------------------|--------------------|------------------|------------------|---------|
| Behaviour with respect to water | hydrophobic | | | | | | | | |
| Appearance | free-flowing white powder | | | | | | | | |
| BET ¹⁾ surface area | m ² /g | 110+20 | 170+20 | 100+20 | 150+25 | 260+30 | 220+25 | 150+25 | 250+30 |
| Average size of primary particles | nm | 16 | 12 | 14 | 12 | 7 | 7 | 12 | 7 |
| Tamped density/ approx. value ²⁾ | | | | | | | | | |
| normal product | g/l | 50 | 50 | 50 | 50 | 50 | 50 | 50 | 50 |
| compacted product (suffix "V") ¹²⁾ | g/l | 90 | 90 | | | | | | |
| Loss on drying ³⁾ (2 hours at 105°C) on leaving supplier's works | % | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | - | - |
| Loss on ignition ^{4) 7)} (2 hours at 1000°C) | % | <2 | <2 | 4-6 | 5-7 | 1.0-2.5 | 1.5-3.0 | - | - |
| C content | % | 0.6-1.2 | 0.7-1.3 | 3.5-5.0 | 4.5-6.5 | 2.0-3.0 | 3.0-4.0 | 1-2 | 1.5-3.0 |
| pH ⁵⁾¹⁰⁾ | | 3.6-4.4 | 3.7-4.7 | 4-6 | 3.5-5.5 | 5.5-7.5 | 5.5-7.5 | >4.0 | >3.7 |

| | | | | | | | | | |
|---|---|-------|-------|--------|--------|--------|--------|--------|--------|
| SiO₂ ⁸⁾ | % | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 | >99.8 |
| AlO₂O₃ ⁸⁾ | % | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 |
| Fe₂O₃ ⁸⁾ | % | <0.01 | <0.01 | <0.01 | <0.01 | <0.01 | <0.01 | <0.01 | <0.01 |
| TiO₂ ⁸⁾ | % | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 | <0.03 |
| HCl ¹¹⁾ | % | <0.05 | <0.1 | <0.025 | <0.025 | <0.025 | <0.025 | <0.002 | <0.025 |

- 1) based on DIN 66131
 2) based on DIN ISO 787/XI, JIS K 5101/18 (not sieved)
 3) based on DIN ISO 787/II, ASTM D 280, JIS K 5101/21
 4) based on DIN 55921, ASTM D 1280, JIS K 5101/23
 5) based on DIN ISO 787/IX, ASTM D 1208, JIS K 5101/24
 7) based on substance dried at 105°C for 2 hours
 8) based on substance ignited at 1000°C for 2 hours
 10) in water:methanol = 1:1
 11) HCl content is a constituent of the loss on ignition
 12) V product is supplied in 15 kg bags

| Properties | | AEROSIL |
|---------------------------------|-------------------|---------------------|
| | | R 8200 |
| Behaviour with respect to water | | hydrophobic |
| BET surface area | m ² /g | 160 ₋ 25 |
| Tamped density | g/l | about 140 |
| Loss on drying (2 h at 105°C) | % | ≤ 0.5 |
| pH (4% in water) | | ≥ 5.5 |
| Carbon content | % | 2.0 - 4.0 |
| SiO ₂ | % | ≥ 99.8 |
| Fe ₂ O ₃ | % | ≤ 0.01 |
| TiO ₂ | % | ≤ 0.03 |
| HCl | % | ≤ 0.025 |

It is preferable that components d) are not zeolites.

15 **Component e)**

The compounds of the formula (I) can by way of example be obtained via a hydrosilylation reaction (reactions of the Si-H-functional siloxanes with allyl polyethers) as described by way of example in EP 1 520 870 A. In particular, the hydrosilylation reaction can be carried out as described in example 1 of that document.

20

The Si-H-functional siloxane to be used for the production of the compounds of the formula (I) can by way of example be produced via equilibration as described in the document EP 1439200 A, in particular in Example 1 of the same document. For the production of siloxanes having terminal hydrogen it is accordingly necessary to use, as raw material, a polymethylhydrosiloxane with terminal hydrogen functionality. The nature and amount of the raw materials here is to be selected in such a way as to give the respective desired siloxane structure.

The allyl polyethers can by way of example be produced by a method based on that described in the document DE 19940797, in particular in accordance with the method described in Example 1 of the said document.

The claimed moulding compositions can comprise water, and in particular the claimed moulding compositions can comprise water when bismuth compounds or bismuth salts are used as catalyst component c). However, preferred claimed moulding compositions comprise less than 2% by weight of water, based on the moulding composition, preferably less than 0.2% by weight of water, and in particular no water. The water does not lead to any foaming of the reaction mixture.

It is preferable that the claimed moulding compositions comprise no components that represent a chemical blowing agent. The expression chemical blowing agents means blowing agents which under the reaction conditions eliminate or produce a gas, with the exception of water. In particular, the claimed moulding composition does not comprise the chemical blowing agent formic acid. In accordance with the present explanation, if any water is present it does not represent a chemical blowing agent.

For the purposes of the present invention, the expression physical blowing agents means compounds which are present in gaseous form at atmospheric pressure and at a temperature below 50°C, preferably below 25°C, and which do not react chemically with any of the other compounds present in the moulding composition.

Preferred physical blowing agents are those selected from the group consisting of carbon dioxide, acetone, hydrocarbons, e.g. n-, iso- or cyclopentane, cyclohexane and halogenated hydrocarbons, e.g. methylene chloride, tetrafluoroethane, pentafluoropropane, heptafluoropropane, pentafluorobutane, hexafluorobutane and dichloromonofluoroethane.

15

As mentioned above, the claimed moulding compositions can comprise physical blowing agents, but in the low concentrations stated above. The density of the resultant claimed mouldings is preferably greater than or equal to 500 kilograms per cubic metre, preferably greater than or equal to 700 kilograms per cubic metre, particularly preferably greater than or equal to 900 kilograms per cubic metre. It is preferable that the claimed moulding compositions comprise no physical blowing agent.

25

The claimed moulding compositions can comprise further constituents alongside the components a) to e) mentioned. The proportion of the said further constituents, based on the total composition, can be up to 75% by weight, based on the total composition. It is preferable that the claimed moulding compositions comprise one or more constituents selected from flame retardant, antioxidants, UV stabilizer, inorganic and/or organic fillers not covered by the definition of component d), colorants, water-binding agents, surfactant substances not covered by the definition

35

of component e), plant-protection agent, extender and/or plasticizer.

Flame retardants used are preferably those that are liquid and/or are soluble in the moulding composition or in one of the other components used. It is preferable to use commercially available phosphorus-containing flame retardants, such as diphenyl cresyl phosphate, tricresyl phosphate, tris(2-chloroethyl) phosphate, tris(2-chloropropyl) phosphate, tris(2,3-dibromopropyl) phosphate, tris(1,3-dichloropropyl) phosphate, tetrakis(2-chloroethyl) ethylenediphosphate, trisbutoxyethyl phosphate, dimethyl methanephosphonate, diethyl ethanephosphonate, diethyl diethanolaminomethylphosphonate. Equally suitable compounds are halogen- and/or phosphorus-containing, flame-retardant polyols and/or melamine, and also expandable graphite. The amount preferably used of the flame retardants is at most 35% by weight, preferably at most 20% by weight, based on the polyol component.

Inorganic fillers comprised in the moulding composition are preferably baryte, chalk, gypsum, kieserite, soda, kaolin, carbon black, metal powder, e.g. iron powder or copper powder and/or glass microbeads.

Examples of organic fillers that can be present in the moulding composition are powders based on polystyrene, polyvinyl chloride, urea-formaldehyde compositions and/or polyhydrazodicarbonamides (e.g. derived from hydrazine and tolylene diisocyanate). It is possible here by way of example that urea-formaldehyde resins or polyhydrazodicarbonamides have been produced directly in a polyol to be used for the claimed moulding composition. It is also possible to add hollow microbeads of organic origin.

35

Inorganic and/or organic fillers can also be present in the form of short fibres. Examples of short fibres that can be used are glass fibres and/or fibres of organic origin, for example polyester fibres or polyamide fibres. The length of the short fibres is preferably from 0.01 to 1 cm.

Examples of colorants that can be present in the claimed moulding compositions are organically and/or inorganically based dyes and/or colour pigments that are known per se for colouring polyurethane, for example iron oxide pigments and/or chromium oxide pigments and phthalocyanine-based and/or monoazo-based pigments, but colorants used comprise no pyrogenic oxide, in particular no pyrogenic oxide selected from silicon dioxide, aluminium oxide, mixed oxide of silicon dioxide and aluminium oxide, titanium dioxide or mixed oxide of titanium dioxide and iron oxide.

Water-binding agents that can preferably be present are zeolites. Suitable synthetic zeolites are available commercially by way of example as Baylith®.

Extenders present in the claimed moulding composition are preferably liquid, practically inert substances with boiling point above 150°C (at atmospheric pressure). Examples that may be mentioned are: alkyl-, alkoxy- and/or halogen-substituted aromatic compounds, such as dodecylbenzene, m-dipropoxybenzene and/or o-dichlorobenzene, halogenated aliphatic compounds such as chlorinated paraffins, organic carbonates such as propylene carbonate, esters of carboxylic acids, for example dioctyl phthalate, and also esters of dodecylsulfonic acid; other examples are organic phosphorus compounds such as tricresyl phosphate.

Examples of plasticizers that can be used in the claimed moulding composition are esters of phosphoric acid or of

polybasic, preferably dibasic carboxylic acids with monohydric alcohols, e.g. esters of succinic acid, isophthalic acid, trimellitic acid, phthalic anhydride, tetra- and/or hexahydrophthalic anhydride, endomethylene-
5 tetrahydrophthalic anhydride, glutaric anhydride, maleic anhydride, fumaric acid and/or dimeric and/or trimeric fatty acids, such as oleic acid, optionally in a mixture with monomeric fatty acids, formed with branched and/or aliphatic alcohols having from 1 to 20 carbon atoms, e.g.
10 methanol, ethanol, propanol, isopropanol, n-butanol, sec-butanol, tert-butanol, the various isomers of pentyl alcohol, of hexyl alcohol, of octyl alcohol (for example 2-ethylhexanol), of nonyl alcohol, of decyl alcohol, of decyl alcohol, of lauryl alcohol, of myristyl alcohol, of cetyl
15 alcohol, stearyl alcohol and/or of fatty and wax alcohols that occur naturally or that are obtainable via hydrogenation of naturally occurring carboxylic acids, or of cycloaliphatic and/or aromatic hydroxyl compounds, such as cyclohexanol and its homologues, phenol, cresol, thymol, carvacrol, benzyl alcohol and/or phenylethanol. It is, of
20 course, also possible to use mixed esters of the abovementioned alcohols and carboxylic acids.

The moulding composition can also preferably comprise
25 phosphoric esters derived from halogenated alcohols, for example trichloroethyl phosphate. These have the advantage that a flame-retardant effect can be achieved at the same time as the plasticizer effect.

30 The plasticizers can also involve "polymeric plasticizers", for example polyesters of adipic, sebacic and/or phthalic acid.

Alkylsulphonic esters of phenol can moreover also be used as plasticizers, examples being phenyl paraffinsulphonates.

The claimed moulding compositions can be used for the production of mouldings. In particular, the claimed moulding compositions can be used in the claimed process described below.

5

The process for the production of mouldings based on reaction products of one or more polyisocyanates and one or more polyols is characterized in that a claimed moulding composition is converted to a moulding by a shaping process, and the reaction for the production of the reaction products at least continues after the shaping process.

In the claimed process, the moulding composition is preferably produced via mixing of the components prior to or during the shaping process. The mixing is preferably achieved by means of a mixing nozzle. The components can be introduced individually, or else in various premixes, to the mixing nozzle. It is preferable that mixtures comprising components b), c) and d) are introduced into the mixing nozzle while each of components a) and e) is introduced separately.

The reaction to give the reaction products begins when components a) to c) are brought into contact. It is therefore preferable that components a) to c) are brought into contact only shortly before or during the shaping process, in such a way that the reaction has not concluded before the moulding is obtained. The conduct of the process is preferably such that when the processing to give the moulding is concluded the conversion achieved in the reaction is at most 50% of the theoretically possible conversion. It is preferable that the components are mixed in a mixing head or static mixer directly before discharge into a mould or onto a surface. In accordance with the claimed process it is possible here by way of example to

produce an appropriately moulded linear product by way of the geometric shape of the discharge nozzle. The residence time in the mixing head or mixer here is preferably less than 20 seconds, with preference less than 10 seconds. The
5 hardening, i.e. the polyurethane reaction, preferably takes place in from 30 sec to 15 min; this can be adjusted via the selection and amount of the catalyst.

It can be advantageous to control the temperature of the
10 reaction mixture. The temperature at which the reactions are then carried out is preferably below 90°C, more preferably below 50°C and in particular preferably below 25°C.

15 In the claimed process it is preferable to use moulding compositions which comprise no blowing agents.

The claimed mouldings can be obtained by means of the claimed process and also through reaction of a moulded
20 claimed moulding composition. The claimed moulding is preferably a gel.

The gels according to the invention have the property of deforming on exposure to a force and, after removal of the
25 deforming force, returning almost completely to their initial state. An effect of this property is that elements which comprise the gels according to the invention can deform under pressure, can conform to surfaces, and can compensate for unevenness. The said property is retained
30 even after repeated application of load to, and removal of load from, the gel.

Components which comprise gels according to the invention can be used in many ways, for example as sealing elements.
35 Direct application as sealing element and/or extensible element with unconstrained geometry is possible according

to the invention. In particular, it is possible to apply the moulding composition to surfaces without any other shaping aids and thus to apply a sealing element, such as a sealing bead, with completely unconstrained geometry.

5 Coating of surfaces with any desired gradients and with vertical planes is likewise possible. By varying component e) it is possible to adjust to a defined thixotropy/viscosity, and this is particularly advantageous for the smooth transition between the start and end of an

10 annular sealing element. To this end, the variation of component e) can take place during metering, in particular in the region of the transition between the beginning and end of the metering of the sealing element.

15 The cross-sectional geometry of the gel can be modified appropriately by using an appropriately shaped diaphragm, i.e. the extruded gel can have, as desired, a round or polygonal cross-sectional geometry or any other type of cross-sectional geometry. Examples of applications here are

20 seals having any desired shapes for housings, in particular seals which are not subject to any spatial restriction and which are not flat, i.e. for which a non-flowable moulding composition is necessary.

25 Another possible application uses gel cushions as vibration-damping element, particularly for solid-borne-sound-deadening. The moulding composition can be applied by spraying, casting, immersion, spreading or other types of process. The adjustable and distinct thixotropy of the

30 claimed moulding composition permits production of geometries and layer thicknesses which would not be obtainable through use of a low-viscosity (or non-thixotropic) moulding composition.

35 In particular, the possible variation of component e) and the resultant variation of viscosity (or thixotropy - i.e.

shear-rate-dependent viscosity) is very advantageous for spraying of the moulding composition. Equally, it is possible to manufacture a sprayed skin with uniform layer thickness in tooling of any desired geometry. By way of example, a gel-type sprayed skin can be used for "soft-touch" elements, for example decorative foils for instrument panels.

The gel according to the invention is adhesive and can therefore be used as adhesion medium, for example as assembly aid for sealing elements.

The claimed moulding is particularly preferably a sealing element, an assembly aid, an adhesion medium, a soft-touch element, a gel cushion or a vibration-damping element, or is particularly preferably used as sealing element, as assembly aid, as adhesion medium, as soft-touch element, as gel cushion or as vibration-damping element.

A typical polyurethane gel formulation for the purposes of this invention would have the following constitution:

Constitution:

| | Component | proportion by weight |
|----|------------------------|----------------------|
| 25 | Polyol | 100 |
| | Pyrogenic oxide | from 1 to 30 |
| | Catalyst | from 0.05 to 1.5 |
| | Siloxane (formula (I)) | from 0.25 to 7.5 |
| | Flame retardant | from 0 to 50 |
| 30 | Isocyanate index: | from 13 to 80 |

The present invention is described by way of example in the examples listed below, but without any intention that the invention, the scope of application of which is apparent from the entire description and from the claims, be restricted to the embodiments specified in the examples.

Examples**Example 1: Production processes for mixtures of components****a), b), c), d) and e)**

5 Mixtures composed of
a) polyisocyanate (only sometimes present)
b) polyol
c) catalyst
d) pyrogenically produced oxide of a metal or of a
10 metalloid
e) organomodified siloxane of the formula (I)
were produced for measurement of thixotropic properties by
technical methods.

15 Addition of polyisocyanates - component a) - causes
hardening of the mixture to begin, and this can make it
more difficult to measure rheological properties. Just a
few measurements with an isocyanate were therefore carried
out, in order to demonstrate retention of the thixotropic
20 properties.

It was also possible to demonstrate thixotropy by using
appropriate experiments in the Examples 3 by casting, onto
a smooth surface, material that does not lose its shape as
25 a result of flow.

Description of components a) to e) used

The following polyisocyanate was used:

No. 1: difunctional HDI prepolymer with 12.5% NCO content,
30 with viscosity 4000 mPa*s

The following polyols were used:

No. 1: trifunctional polyalkylene glycol composed of EO and
PO with 82 mol% content of PO, with OH number 35 mg KOH/g
35 and with viscosity 860 mPa*s

No. 2: trifunctional Polyalkylene glycol composed of PO with OH number 56 mg KOH/g and with viscosity 660 mPa*s, obtainable as Arcol polyol 1104 from Bayer MaterialScience

5 No. 3: trifunctional polyalkylene glycol composed of EO and PO with 85 mol% content of PO, OH number = 32 mg KOH/g and with viscosity 1390 mPa*s obtainable as Hyperlite 1629 from Bayer MaterialScience

10 No. 4: trifunctional polyalkylene glycol composed of PO with OH number 56 mg KOH/g and with viscosity 550 mPa*s, obtainable as Voranol CP 3322 from Dow

The following catalyst was used:

No. 1: bismuth tris(neodecanoate) - COSCAT 83[®] from Erbslöh

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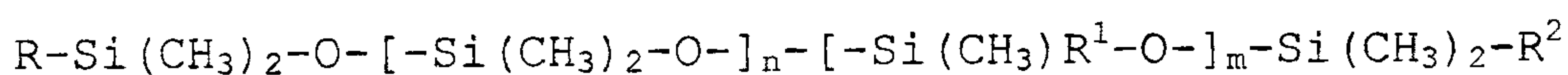
The following pyrogenic oxides were used:

No. 1: Aerosil[®] R 8200 from Evonik Industries AG

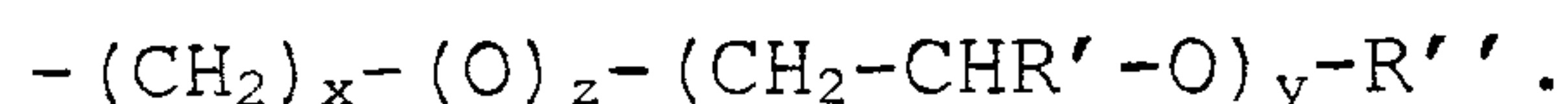
No. 2: Aerosil[®] R 805 from Evonik Industries AG

20 The following siloxanes were used:

Organomodified siloxanes of the formula (I)



25 having moieties R¹ of the formula (II)



30 As described in Example 1 of document EP 1439200 A1, the Si-H-functional siloxanes to be used were produced from the corresponding siloxane raw materials via equilibration. (For the production of siloxanes with terminal modification, a polymethylhydrosiloxane with terminal hydrogen functionality must accordingly be used as raw material.) The nature and amount of the raw materials was

35

selected in such a way as to give the respective desired siloxane structure.

The allyl polyethers were produced by analogy with the method described in Example 1 of DE 19940797 A1, in this case using allyl alcohol as starter and, as appropriate, ethylene oxide and propylene oxide or styrene oxide.

The hydrosilylation reactions (reactions of the Si-H-functional siloxanes with the allyl polyethers) were carried out in accordance with Example 1 in document EP 1520870 A1.

Table 1 collates the structures used for the moieties R¹. Table 2 describes the siloxanes according to the invention. The terminology and indices in formula (I) were used. All % data in Table 1 and Table 2 are in mol%. The abbreviation Me in Tables 1 and 2 means a methyl group.

Table 1: Description of side chains R¹:

| Name | R' | R'' | x | y | z |
|------|-----------------------------|-----|---|----|---|
| A | 35 mol% of Me; 65 mol% of H | H | 3 | 23 | 1 |
| B | 20 mol% of Me; 80 mol% of H | H | 3 | 15 | 1 |
| C | 100 mol% of H | H | 3 | 12 | 1 |
| D | 20 mol% of Me; 80 mol% of H | H | 3 | 13 | 1 |
| E | 100 mol% of H | H | 3 | 8 | 1 |
| F | - | H | 6 | 0 | 1 |
| G | 100 mol% of H | H | 3 | 1 | 1 |
| H | 100 mol% of H | Me | 3 | 8 | 1 |

Table 2: Description of siloxanes Nos. 1 to 17:

| Siloxane | R | R ² | R ¹ | n | m |
|----------|----------------|----------------|----------------|----|----|
| 1 | Me | Me | 100 mol% of E | 35 | 3 |
| 2 | Me | Me | 100 mol% of C | 28 | 10 |
| 3 | R ¹ | R ¹ | 100 mol% of A | 40 | 3 |
| 4 | Me | Me | 100 mol% of C | 20 | 2 |
| 5 | Me | Me | 100 mol% of B | 50 | 7 |
| 6 | Me | Me | 100 mol% of E | 60 | 7 |
| 7 | R ¹ | R ¹ | 100 mol% of C | 17 | 1 |
| 8 | R ¹ | R ¹ | 100 mol% of E | 75 | 1 |
| 9 | Me | Me | 100 mol% of E | 75 | 5 |
| 10 | Me | Me | 100 mol% of D | 35 | 3 |
| 11 | R ¹ | R ¹ | 100 mol% of F | 9 | 0 |
| 12 | R ¹ | R ¹ | 100 mol% of E | 35 | 3 |
| 13 | Me | Me | 100 mol% of E | 65 | 13 |
| 14 | Me | Me | 100 mol% of E | 40 | 10 |
| 15 | Me | Me | 100 mol% of G | 40 | 3 |
| 16 | R1 | R1 | 100 mol% of H | 35 | 3 |
| 17 | Me | Me | 100 mol% of H | 60 | 7 |

Various mixtures were produced via mixing of the appropriate starting materials. The Aerosil grades here were stirred into the appropriate polyols with the aid of a disperser disc at rotation rates of 500 to 2000 rpm. The other components (catalyst, siloxanes and optionally isocyanate) were then stirred into the mixture at lower rotation rates of from 50 to 500.

10

The amounts used of the starting materials can be found in Table 3. All of the mixtures specified comprised 0.2 part of bismuth trisneodecanoate, based on 100 parts of polyol.

Table 3: Constitution of the mixtures in proportions by mass

| Ex. No. | Polyol | | Pyrogenic oxide | | Siloxane | |
|--------------|--------|-------|-----------------|-------|----------|-------|
| | No. | Parts | No. | Parts | No. | Parts |
| 1.comp.1 | 1 | 100 | 1 | 15 | - | |
| 1.1 | 1 | 100 | 1 | 15 | 6 | 3 |
| 1.2 | 1 | 100 | 1 | 15 | 6 | 0.5 |
| 1.3 | 1 | 100 | 1 | 15 | 12 | 0.5 |
| 1.4 | 1 | 100 | 1 | 15 | 6 | 1 |
| | | | | | | |
| 1. comp.2 | 1 | 100 | 2 | 10 | - | |
| 1.5 | 1 | 100 | 2 | 10 | 6 | 1 |
| 1.6 | 1 | 100 | 2 | 10 | 8 | 1 |
| 1.7 | 1 | 100 | 2 | 10 | 9 | 1 |
| 1.8 | 1 | 100 | 2 | 10 | 12 | 1 |
| | | | | | | |
| 1. comp. 3 | 4 | 100 | 2 | 10 | | |
| 1.9 | 4 | 100 | 2 | 10 | 6 | 1 |
| 1.10 | 4 | 100 | 2 | 10 | 4 | 1 |
| 1.11 | 4 | 100 | 2 | 10 | 5 | 1 |
| 1.12 | 4 | 100 | 2 | 10 | 3 | 1 |
| | | | | | | |
| 1. comp. 4 | 3 | 100 | 2 | 10 | | |
| 1.13 | 3 | 100 | 2 | 10 | 1 | 1 |
| 1.14 | 3 | 100 | 2 | 10 | 2 | 1 |
| 1.15 | 3 | 100 | 2 | 10 | 7 | 1 |
| 1.16 | 3 | 100 | 2 | 10 | 6 | 1 |
| | | | | | | |
| 1. comp. 5 | 2 | 100 | 1 | 15 | | |
| 1.17 | 2 | 100 | 1 | 15 | 6 | 3 |
| 1.18 | 2 | 100 | 1 | 15 | 6 | 2 |
| 1.19 | 2 | 100 | 1 | 15 | 6 | 1 |
| 1.20 | 2 | 100 | 1 | 15 | 5 | 1 |
| 1.21 | 2 | 100 | 1 | 15 | 10 | 1 |
| 1.22 | 2 | 100 | 1 | 15 | 4 | 1 |
| 1.23 | 2 | 100 | 1 | 15 | 3 | 1 |
| 1.24 | 2 | 100 | 1 | 15 | 11 | 1 |
| 1.25 | 2 | 100 | 1 | 15 | 13 | 1 |
| 1.26 | 2 | 100 | 1 | 15 | 14 | 1 |
| 1.27 | 2 | 100 | 1 | 15 | 15 | 1 |
| 1.28 | 2 | 100 | 1 | 15 | 16 | 1 |
| 1.29 | 2 | 100 | 1 | 15 | 17 | 1 |
| | | | | | | |
| | | | | | | |
| 1. comp.6 *) | 1 | 100 | 1 | 15 | - | |
| 1.30*) | 1 | 100 | 1 | 15 | 6 | 2 |
| 1.31*) | 1 | 100 | 1 | 15 | 6 | 3 |
| | | | | | | |
| | | | | | | |

*) 12 parts of isocyanate No. 1 were also present here

Example 2: Determination of the thixotropic properties of the mixtures

The measurements were carried out by using an Anton Paar MCR 301 rheometer with plate-on-plate (40 mm) geometry at a temperature of 25°C. The following were determined for the mixtures produced as in the example: storage modulus, loss modulus, yield point, and yield point recovery times. The samples were tested in oscillation at from 0.1 to 200 Pa (20 points, 1 Hz). The storage modulus and loss modulus were determined from the plateau values of the linear viscoelastic (LVE) region. The yield point is the shear stress at which the storage modulus deviates from ideal viscoelastic behaviour.

Recovery after shear was determined by testing the sample for 100 seconds (20 × 5 seconds) at 1 Pa (1 Hz). The shear stress was then increased to 500 Pa (1 Hz) for 100 seconds (20 × 5) before again testing at 1 Pa (1 Hz). The recovery time is determined from the time required for the storage modulus to be greater than the loss modulus. Table 4 collates the results of the tests of Example 2.

Table 4: Rheological properties of mixtures of Example 1

| Mixture | Storage modulus / Pa | Loss modulus / Pa | Yield point / Pa | Recovery time / s |
|-----------|----------------------|-------------------|------------------|-------------------|
| 1.comp.1 | 0.1 | 6.1 | - | - |
| 1.1 | 9290 | 4460 | 12.2 | 0 |
| 1.2 | 1030 | 445 | 5.5 | 15 |
| 1.3 | 383 | 209 | 2.5 | 40 |
| 1.4 | 4370 | 1610 | 12.2 | 0 |
| | | | | |
| 1.comp. 2 | 52 | 72.8 | - | - |
| 1.5 | 1630 | 718 | 1.7 | 0 |
| 1.6 | 675 | 311 | 2.5 | 0 |
| 1.7 | 1880 | 584 | 1.7 | 0 |
| 1.8 | 3400 | 927 | 3.7 | 0 |
| | | | | |
| 1.comp. 3 | 2930 | 733 | 2.5 | 0 |
| 1.9 | 8820 | 1500 | 18.1 | 0 |
| 1.10 | 7290 | 993 | 8.1 | 0 |
| 1.11 | 7900 | 1370 | 3.7 | 5 |
| 1.12 | 11800 | 2120 | 12.2 | 0 |
| | | | | |
| 1.comp. 4 | 46.4 | 65.4 | | |
| 1.13 | 2080 | 600 | 1.7 | 0 |
| 1.14 | 1410 | 500 | 1.7 | 0 |
| 1.15 | 303 | 212 | 2.5 | 5 |
| 1.16 | 3860 | 820 | 8.2 | 0 |
| | | | | |
| 1.comp. 5 | 22.7 | 36 | - | - |
| 1.17 | 14700 | 3110 | 15.7 | 0 |
| 1.18 | 18700 | 4020 | 11.5 | 0 |
| 1.19 | 15600 | 3210 | 15.7 | 0 |
| 1.20 | 2450 | 965 | 6.1 | 5 |
| 1.21 | 377 | 204 | 4.5 | 15 |
| 1.22 | 5160 | 1590 | 11.5 | 0 |
| 1.23 | 396 | 247 | 4.5 | 20 |
| 1.24 | 685 | 316 | 2.5 | 0 |
| 1.25 | 3550 | 1042 | 4.5 | 0 |
| 1.26 | 3120 | 867 | 3.7 | 0 |
| 1.27 | 715 | 345 | 2.5 | 0 |
| 1.28 | 1340 | 626 | 4.5 | 0 |
| 1.29 | 4200 | 190 | 6.1 | 0 |
| | | | | |
| | | | | |
| 1.comp.6 | 3.7 | 16.7 | | |
| 1.30 | 1790 | 826 | 5.7 | 0 |
| 1.31 | 4040 | 1450 | 8.7 | 0 |

The tests confirm that addition of the siloxanes render the
5 mixtures thixotropic or, as in Examples 1.comp.3, 1.9,
1.10, 1.11 and 1.12, rendered a composition more

thixotropic. It is therefore possible to avoid any relatively high level of Aerosil fill and nevertheless to raise the yield point.

5 In the other examples, thixotropy was achieved only by addition of the siloxanes.

Example 3: Production of PU gels

**Production of the combination of components b), c) and d)
10 (polyol mixture):**

Component b) was used as initial charge. Component c) was incorporated homogeneously by mixing with a propeller stirrer at a rotation rate of 500 rpm. Component d) was then dispersed by means of a disperser disc at a rotation
15 rate of 3000 rpm for 5 minutes. The resultant polyol mixture was devolatilized for 2 hours at subatmospheric pressure (0.2 bar absolute).

**Production of the combination of polyol mixture and
20 components a) and e) (moulding composition):**

A low-pressure three-component metering system was used to convey and mix the polyol mixture, component a) and component e). Homogenization of the individual components was achieved by using a 13-12-SR-POM static-dynamic mixing
25 tube, mixer rotation rate = 4000 rpm, mass flow rate about 600 g/min. The metering of component e) into the system could be switched on or off as desired.

For the experiments Example 3.1 and Example 3.2 specified
30 below, the compositions of Example No. 1.31 and 1.comp.6 from Table 3 were selected. For comparison, in each case 10 mL of the moulding compositions were spot-applied to a smooth substrate. The diameters of the applied spots of moulding compositions were determined directly after
35 application and 30 seconds after application. The effect of component e) on the elongation at break and tensile

strength property of the material was also tested on compositions of Examples Nos. 1.31 and 1.comp.6 (ISO 53504 - S2 test specimen, thickness of material 3 mm).

5 **Example 3.1 (not according to the invention)**

Application of a moulding composition corresponding to the composition of Example 1.comp.6 gives a spot of moulding composition of diameter 6 cm, and after 30 seconds 9 cm, i.e. the moulding composition flows. The elongation at
10 break determined was 1251%, and tensile strength was 1.68 MPa.

Example 3.2 (according to the invention)

Application of a moulding composition corresponding to the
15 composition of Example 1.31 gives a spot of moulding composition of diameter 4 cm, and after 30 seconds 4 cm, i.e. the moulding composition does not flow. The elongation at break determined was 1198%, and tensile strength was 1.52 MPa.

20

Addition of component e) (Example 3.2) produces a yield point (Table 4). In a commercial application, the moulding composition can therefore be applied directly in the form of moulding, since it does not lose its shape of a result
25 of flow. This means in practice that the claimed moulding composition can be applied directly in the form of moulding whereas moulding compositions not according to the invention lose their shape immediately after application.

30 Addition of component e) did not significantly affect the mechanical properties of the gels. The changes amount to <10%, based on the initial values of the properties, i.e. the improvement described in EP 1125975 A1 in the properties of the gel is approximately retained.

35

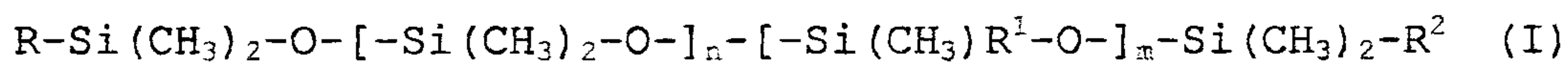
The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A moulding composition comprising the following components:

- a) at least one polyisocyanate;
- b) at least one polyol;
- c) at least one catalyst catalysing the reaction between the polyisocyanate and polyol; and
- d) at least one pyrogenically produced oxide of a metal or of a metalloid;

wherein the moulding composition comprises, as further component;

e) at least one siloxane of the formula (I)



where

R, R¹ and R² are identical or different;

R and/or R² are methyl or R¹;

wherein R¹ at each occurrence is:

-(CH₂)_x-(O)_z-(CH₂-CHR'-O)_y-R''',

-(CH₂)-R''',

-(CH₂)_x-(O)_z-R''',

-CH₂-CH₂-CH₂-O-CH₂-CH(OH)-CH₂OH, or

-(CH₂)_x-(O)_z-(CH₂-CHR'-O)_y-R''';

wherein R' at each occurrence is -H, -CH₃, -CH₂CH₃, or a phenyl moiety;

wherein R'' at each occurrence is -H, -(CO)-R''', -(CO)-NH-R''' or C₁ to C₄₀-alkyl;

wherein R''' at each occurrence is C₁ to C₄₀-alkyl, -aryl or -alkylaryl; optionally also substituted with at least one halogen;

wherein n+m+2 = 10 to 150;

m = 0 to 20,

x = 2 to 15,

y = 1 to 40,

z = 0 or 1,

wherein the units (CH₂-CHR'-O) are identical or different;

with the proviso that for m = 0 at least one moiety R or R² is identical with R¹;

with the proviso that if z=0, x and y = 0 and R'' has at least 2 carbon atoms; and

with the proviso that the moulding composition comprises less than 2% by weight, based on the entire constitution of the moulding composition, of physical blowing agent.

2. A moulding composition according to Claim 1, wherein R'' is C₁- or C₆- to C₃₀-alkyl.
3. A moulding composition according to Claim 1 or 2, wherein n+m+2= 12 to 85.
4. A moulding composition according to Claim 1 or 2, wherein n+m+2= 15 to 47.
5. A moulding composition according to any one of Claims 1 to 4, with the proviso that the moulding composition comprises less than 0.2% by weight based on the entire constitution of the moulding composition, of physical blowing agent.
6. A moulding composition according to any one of Claims 1 to 5, wherein m = 1 to 4.
7. A moulding composition according to any one of Claims 1 to 6, wherein x = 3 to 10.

8. A moulding composition according to any one of Claims 1 to 7, wherein $y = 2$ to 19.
9. A moulding composition according to any one of Claims 1 to 8, wherein in the at least one siloxane compound of the formula (I) used as component e), the moieties R^1 are $-(CH_2)_x-(O)_z-(CH_2-CHR'-O)_y-R''$ moieties, where R' , R'' , x , y , and z are as defined in any one of Claims 1 to 8.
10. A moulding composition according to Claim 9, wherein in the at least one siloxane compound of the formula (I) present as component e), the numeric-average molar proportion of alkylene oxide units where $R'=H$, based on all of the alkylene oxide units in the moieties R^1 , is at least 80%.
11. A moulding composition according to Claim 9, wherein in the at least one siloxane compound of the formula (I) present as component e), the numeric-average molar proportion of alkylene oxide units where $R'=H$, based on all of the alkylene oxide units in the moieties R^1 , is at least 90%.
12. A moulding composition according to Claim 9, 10 or 11, wherein in the at least one siloxane compound of the formula (I) present as component e), at least 80% of the moieties R'' are hydrogen.
13. A moulding composition according to any one of Claims 1 to 12, wherein in the at least one siloxane compound of the formula (I) present as component e), the quotient $Q = \text{number of Si atoms}/\text{number of moieties } R^1$ (in each case in numeric average per molecule) is from more than 5 to less than or equal to 16.

14. A moulding composition according to any one of Claims 1 to 12, wherein in the at least one siloxane compound of the formula (I) present as component e), the quotient $Q = \text{number of Si atoms} / \text{number of moieties } R^1$ (in each case in numeric average per molecule) is from more than 8 to less than or equal to 12.
15. A moulding composition according to any one of Claims 1 to 14, wherein bismuth trisneodecanoate is present as component c).
16. A moulding composition according to any one of Claims 1 to 15, wherein the proportion of component d) in the moulding composition is from 1 to 30 parts by weight, per 100 parts by weight of component b).
17. A moulding composition according to any one of Claims 1 to 15, wherein the proportion of component d) in the moulding composition is from 5 to 20 parts by weight, per 100 parts by weight of component b).
18. A moulding composition according to any one of Claims 1 to 17, wherein the proportion of component e) in the moulding composition is from 0.05 to 5 parts by weight, per 100 parts by weight of component b).
19. A moulding composition according to any one of Claims 1 to 17, wherein the proportion of component e) in the moulding composition is from 0.2 to 5 parts by weight, per 100 parts by weight of component b).
20. A process for the production of a moulding based on reaction products of at least one polyisocyanate and at least one polyol, wherein a moulding composition as

defined in any one of Claims 1 to 19 is converted to a moulding by a shaping process, and the reaction for the production of the reaction products at least continues after the shaping process.

21. A process according to Claim 20, wherein the moulding composition is produced via mixing of the components prior to or during the shaping process.
22. A process according to Claim 20 or 21, wherein a moulding composition comprising no blowing agent is used.
23. A moulding obtained via a process as defined in any one of Claims 20 to 22, where the moulding is a gel.
24. A moulding obtained via reaction of a moulded moulding composition as defined in any one of Claims 1 to 19.
25. A moulding according to Claim 23 or 24, wherein the moulding is a gel.
26. A moulding according to any one of Claims 23 to 25, wherein the moulding is a sealing element, an assembly aid, an adhesion medium, a soft-touch element, a gel cushion or a vibration-damping element.
27. Use of a moulding as defined in any one of Claims 23 to 25 as sealing element, as assembly aid, as adhesion medium, as soft-touch element, as gel cushion or as vibration-damping element.