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(54) **Title:** MOLDING-COMPOSITIONS COMPOSED OF FILLER-REINFORCED THERMOPLASTIC MATERIAL WITH VERY GOOD SCRATCH RESISTANCE AND SOFT-TOUCH FEEL

(57) **Abstract:** The invention relates to a polymeric molding composition composed of a soft material, of a glassy material as filler, and of a thermoplastic polymer. The polymeric molding composition is suitable for the preparation of moldings with good stiffness, with high scratch resistance and with a highly pleasant soft-touch feel. Such moldings are used in the automotive industry or else suitable for household devices.

# Molding compositions composed of filler-reinforced thermoplastic material with very good scratch resistance and soft-touch feel

The invention relates to polymeric molding compositions composed of a soft material, of a filler, and of a thermoplastic polymer, preferably an olefin polymer. The invention relates further to a process for the preparation of such molding composition by bringing the components together and mixing them, and to the use of such molding composition for the production of moldings with low to high stiffness, with good scratch resistance, and with pleasant soft-touch feel, and also to the use of such moldings.

For the purposes of the present application, "soft-touch feel" is a property of the moldings which can be described as a softness perceived as pleasant on contact of the human hand with the surface of the moldings. "Soft-touch feel" is qualitatively determined via a panel composed of five persons of which at least three have to assess the contacted surface as having typical "soft-touch feel" in comparison with an internal sample catalog composed of five different surfaces with and without "soft-touch feel".

The reinforcement of plastic by means of glass fibers has been known for a long time period. By way of example, EP-A 663 418 describes a polypropylene molding composition reinforced with glass fibers. However, achievement of good mechanical properties is attended by an unpleasant hardness of the material, felt by the hand on touching the surface of moldings produced therefrom.

An object underlying the present invention was therefore to provide a thermoplastic molding composition which exhibits good mechanical properties and at the same time has a pleasant soft-touch feel and in addition thereto maintains a high scratch resistance.

Surprisingly, it was found that this object is achieved by the molding composition as mentioned at the outset, which comprises at least a combination of from 5 to 90 % by weight of soft material with from 5 to 60 % by weight of glass material as filler and with from 3 to 70 % by weight of a thermoplastic polymer. The molding composition according to the invention preferably has a melt index $MI_{(230/2.16)}$ in the range of from 0.1 to 100 g/10 min and a tensile modulus in the range of from 10 to 4500 MPa preferably from 100 to 4000 MPa more preferred from 130 to 2500 MPa.

It is an essential feature of the invention that the molding composition comprises from 5 to 90 % by weight, preferably from 10 to 80 % by weight, of soft material. According to the invention, the soft material used can comprise crosslinked or uncrosslinked ethylene-propylene copolymers whose ethylene content is greater than 20 % by weight, or else crosslinked or uncrosslinked ethylene-butene copolymers, or else crosslinked or uncrosslinked ethylene-hexene copolymers, or else crosslinked or uncrosslinked ethylene-propylene-diene-copolymer (EPDM), or else crosslinked or uncrosslinked synthetic or natural rubbers, or else crosslinked or uncrosslinked styrene-ethylene-butadiene terpolymers (SEBS), or else crosslinked or uncrosslinked polybutylene, or else PVC. The particularly preferred amount of soft material of the above type in the molding composition of this invention is in the range of from 10 to 70 % by weight, based on the total weight of the molding composition.

Another essential feature of the inventive polyolefin molding composition is a content of from 5 to 60 % by weight of glass material as filler, based on the total weight of the molding composition. The glass material can be spun glass fibers or can be chopped spun glass fibers whose average length is from about 100 µm to 10 mm, or can be glass particles in the form of ground glass or milled glass fibers whose average particle size is in the range from 3 µm to 5 mm, or glass beads whose average bead diameter is in the range from 3 µm to 2 mm, the average length of the glass fibers in the inventive molding composition being in the range from 200 µm to 5 mm, or the average particle size of the glass particles in the inventive molding composition in the form of ground glass or milled glass fibers being in the range from 5 µm to 2 mm, or the average bead diameter of the glass beads in the inventive molding composition being in the range from 5 µm to 2 mm. According to the invention a further preference is that from 5 to 50 % by weight of spun glass fibers are used, particularly preferably from 10 to 40 % by weight. If appropriate, also a combination with other manifestations of glass as described above and/or other mineral fillers is possible.

Another essential feature of the molding composition is the content of an amount of from 3 to 70 % by weight of thermoplastic polymer in the inventive molding composition, based on the total weight of the molding composition, preferably from 5 to 50 % by weight. The thermoplastic polymer used may be propylene homopolymer or propylene-ethylene copolymer whose ethylene content is smaller than 20 % by weight, or else polyamide, polyester, polystyrene, polycarbonate, or acrylonitrile-butadiene-styrene terpolymer (ABS), PVC, or the like. According to the invention, preference is given to propylene homo- or

copolymer. The particularly preferred amounts used of this copolymer being from 10 to 40 % by weight. Very particular preference is given to the use of propylene polymer and in particular use is made here of a propylene homopolymer or a propylene random copolymer having up to 20 % by weight of copolymerized other olefins having up to 10 carbon atoms. These other olefins are in particular $C_2$-$C_{10}$ 1-alkenes, such as ethylene, 1-butene, 1-pentene, 1-hexene, 1-heptene or 1-octene, preference being given to use of ethylene, 1-butene, or ethylene and 1-butene. The use of propylene homopolymer is particularly preferred.

It is preferable to use an isotactic polypropylene whose fraction insoluble in xylene is above 95 %, preferably above 97 %. The MFR $_{(230\,°C\,/\,2.16\,kg)}$ of the polyolefin is generally from 0.1 to 2500 g/10 min to ISO 1133, preferably from 0.2 to 100 g/10 min, and particularly preferably from 0.3 to 30 g/10 min. Polypropylene molding compositions with monomodal molar mass distribution are also preferred.

To couple the glassy fillers to the polymer matrix, it can be advantageous to use a polar-functionalized compatibilizer. To this end, use can be made of low-molecular-weight compounds which serve to make the glass component less hydrophilic and therefore more compatible with the polymer. Examples of suitable compounds are peroxides, such as tert.-butyl peroxide, or silanes, such as aminosilanes, epoxysilanes, amidosilanes, acrylic silanes, or maleic-anhydride-grafted polyolefins.

In practice it can also be advantageous for the inventive molding composition also to receive additions of stabilizers with respect to heat, stabilizers with respect to UV radiation, or stabilizers with respect to oxidative degradation, or of pigments, or of antistatic agents, or of flame retardants, or of other known additives, each in amounts respectively usual for these additions. The usual amounts of additions of this type are somewhere in the range from 0.1 to 10 % by weight.

The combination of soft material, glassy filler, and thermoplastic gives a molding composition improved over the prior art in relation to the scratch resistance of the moldings produced therefrom. The stiffness of the moldings is expressed in a modulus of elasticity measured to DIN EN ISO 527 or DIN EN ISO 178 in the preferred range from 10 to 4500 MPa. Surprisingly, furthermore, despite the presence of glass as filler, a pleasant "soft-touch feel" is found on the moldings composed of the inventive molding composition, and is qualitatively determined

via a panel composed of five persons of which at least three have to assess the contacted surface as having typical "soft-touch feel" in comparison with an internal sample catalog composed of five different surfaces with and without "soft-touch feel". Another entirely surprising feature is improved scratch resistance of the surface of the moldings composed of the inventive molding composition, expressed qualitatively in dL* values of at most |1.0| using a force of 10 N, measured along to the Volkswagen AG test specification PV 3952.

The inventive molding compositions are obtainable via melting and mixing of the individual components, the mixing process using a mixing apparatus at temperatures of from 120 to 400 °C, preferably from 180 to 280 °C, particularly preferably from 200 to 260 °C, in the presence or absence of peroxides. Mixing apparatus which can be used here are in particular extruders or kneaders, particularly preferably twin-screw extruders. It can also be advantageous to premix the soft material with the thermoplastic polymers and, if appropriate, with other additives at room temperature in a mixing apparatus. If the glassy fillers used comprise long spun glass fibers, this permits advantageous use of the known pultrusion technique in which the spun glass fibers are coated with the soft material and with the thermoplastic polymer in molten form and extruded through dies. Finally, it is also possible to mix the components in suitable premixes in the machines for production of the finished parts or semifinished products (e.g. injection molding machines).

The inventive molding compositions can moreover comprise conventional additives and auxiliaries, such as stabilizers with respect to harmful effects of processing, with respect to thermal oxidation, ageing, effects of UV, neutralizing agents, antistatic agents, waxes or specific low-molecular-weight lubricants, or organic and inorganic pigments and, respectively, pigment preparations, such as carbon black dispersions in polyolefins. The amount of additives and auxiliaries should, however, not exceed 10 % by weight, based on the total amount of the material, preferably 5 % by weight. For optical reasons it can also be advisable to use addition of appropriate suitable colored pigments to the inventive molding compositions to pigment these to give a different, preferably darker, color.

The thermoplastic polyolefins used in the inventive molding compositions may be obtained using any of the familiar processes and catalysts. Preference is given to polymerization of the appropriate monomers by means of a Ziegler-Natta catalyst, of a Phillips catalyst based on chromium oxide, or of a metallocene catalyst. Metallocenes here are intended to mean complexes composed of metals of

groups 3 to 12 of the Periodic Table of the Elements using organic ligands, where these together with metallocenium-ion-forming compounds give effective catalyst systems.

To prepare the olefin polymers, use may be made of the conventional reactors used for polymerization of $C_2$–$C_{10}$ olefins. Suitable reactors are inter alia continuously operated horizontal or vertical stirred tanks, circulating reactors, loop reactors, multistage reactors, or fluidized-bed reactors. The size of the reactors is not significant for preparation of the inventive molding compositions. It depends on the output intended to be achieved in the reaction zone or in the individual reaction zones. The polymerization process may be conducted in one or more stages. The polymerization can be carried out in the gas phase, in bulk or suspension, or in a combination thereof.

The inventive molding composition is particularly suitable for the production of automotive vehicle parts which are subject to stringent requirements for the surface quality of the material. Use in dark-pigmented motor vehicle parts or those provided with a decorative layer is particularly advantageous, examples being center consoles or dashboard cladding. The surface softness of moldings produced from the inventive molding composition makes them suitable, even when chopped spun glass fibers are used, for high-quality applications which were previously accessible only to other molding compositions.

**Working examples 1 to 6**

Various mixtures composed of the soft material SOFTELL CA 02 A, a rubbery $C_2$-$C_3$ copolymer having a $C_2$ content of about 40 % and a $C_3$ content of about 60 %, obtainable from Basell in Italy, with chopped glass fibers whose average length is 4.5 mm, obtainable as ECS O3T 480 from Nippon Electric Glass Co., Ltd., and with thermoplastic polypropylene obtainable as MOPLEN HF501N from Basell in Italy and having an $MI_{(230/2.16)}$ of 12 g/10 min are prepared in a ZSK 53 twin-screw extruder from Coperion-Werner&Pfleiderer at a temperature of 230 °C. Di-tert-butyl peroxide and finally ground talc whose average grain size was 1.4 µm were also added in some inventive examples, the grain size of the talc being determined by the Sedigraph method.

Table 1 below gives the precise constitution of the mixtures.

Table 1

| Working example | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| Soft mat. | 69.5 % | 59.5 % | 70 % | 60 % | 68.5 % | 58.5 % |
| Glass particles | 20 % | 20 % | --- | --- | --- | --- |
| Glass fibers | --- | ---- | 20 % | 20 % | 20 % | 20 % |
| Thermoplastic Polypropylene | 10 % | 20 % | 10 % | 20 % | 10 % | 20 % |
| Peroxide | 0.5 % | 0.5 % | --- | --- | 0.5 % | 0.5 % |
| Talc | --- | --- | --- | --- | 1 % | 1 % |

The mixtures pursuant to the working examples 1 through 6 were used to produce test specimens in accordance with the standard DIN EN ISO 294-1. The test specimens were used to determine the following features:

+ $MI_{230/2.16}$ in [g/10 min] to DIN EN ISO 1133

+ Tensile modulus in [MPa] to DIN EN ISO 527/1+2

+ Shore hardness A 3s to DIN EN ISO 868

+ Shore hardness D 3s to DIN EN ISO 868

+ Scratch resistance dL* to the Volkswagen AG test specification VW PV 3952 on sample sheets with "K-85" surface grain using a loading weight of 10 N.

+ Soft-touch feel measured by the comparison method explained at the outset, in which the meanings of the symbols are as follows: (+) means

good soft-touch feel, (±) means moderate soft-touch feel, and (-) means poor soft-touch feel.

The results of the inventive examples 1 to 6 are illustrated in table 2 below

Table 2

| Working example | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| $MI_{230/2.16}$ | 5.98 | 7.55 | 0.51 | 0.83 | 6.32 | 9.16 |
| Tensile modulus | 139 | 188 | 561 | 1089 | 668 | 1555 |
| Shore hardness A 3s | 85 | 88 | 82 | 89 | 88 | 90 |
| Shore hardness D 3s | 30 | 36 | 37 | 45 | 37 | 48 |
| Scratch resistance | - 0.4 | 0.1 | -0.3 | -0.1 | -0.1 | 0.03 |
| Soft-touch feel | (+) | (+) | (+) | (+) | (+) | (+) |

Comparative examples 1 to 6

For comparison, other mixtures in which one of the inventively used components was omitted were prepared in the ZSK 53 twin-screw extruder used in working examples 1 to 6 under the same conditions, their precise constitutions being identified in table 3 below.

Table 3

| Comparison | CE 1 | CE 2 | CE 3 | CE 4 | CE 5 | CE 6 |
|---|---|---|---|---|---|---|
| Soft mat. | 80 % | 70 % | 60 % | 70 % | 89.5 % | 79 % |
| Glass particles | --- | --- | --- | --- | 10 % | 20 % |
| Wollastonite | 20 % | 20 % | 20 % | 29 % | --- | --- |
| Thermoplastic Polypropylene | --- | 10 % | 20 % | --- | --- | --- |
| Peroxide | --- | --- | --- | --- | 0.5 % | 1 % |
| Talc | --- | --- | --- | 1 % | --- | --- |

Specimens were then produced as in working examples 1 to 6 from the mixtures of comparative examples CE 1 to CE 6. The test specimens were then used with the same test methods to determine features which were the same as those determined in working examples 1 to 6.

The results of the measurements are illustrated in table 4 below.

Table 4

| Comparison | CE 1 | CE 2 | CE 3 | CE 4 | CE 5 | CE 6 |
|---|---|---|---|---|---|---|
| $MI_{230/2.16}$ | 5.9 | 6.64 | 7.62 | 8.72 | 6.41 | 5.6 |
| Tensile modulus | 50 | 123 | 269 | 58 | 32 | 49 |
| Shore hardness A 3s | 75 | 82 | 86 | 77 | 75 | 78 |
| Shore hardness D 3s | 19 | 27 | 36 | 21 | 20 | 22 |
| Scratch resistance | 1.9 | 3 | 2.3 | 3.4 | -2 | -1.4 |
| Soft-touch feel | (+) | (±) | (-) | (±) | (+) | (+) |

From the inventive examples in comparison with the comparative examples, it becomes clear that the inventive combination of a soft material with a glassy filler and with a thermoplastic polymer achieves very good scratch resistance and a pleasant soft-touch feel. If, by way of example, the molding composition does not comprise the thermoplastic polymer, or if, by way of example, a mineral filler, such as Wollastonite, is used to replace the glass material, the scratch resistance of the sample sheets is poor.

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## Patent claims

1. A polymeric molding composition for production of self-supporting moldings with good stiffness, with high scratch resistance, and with pleasant soft-touch feel, which polymeric molding composition comprises at least a combination of from 5 to 90 % by weight of soft material with from 5 to 60 % by weight of glass material as filler and with from 3 to 70 % by weight of a thermoplastic polymer.

2. The polymeric molding composition according to claim 1, which has a melt index $MI_{(230/2.16)}$ in the range of from 0.1 to 100 g/10 min and a tensile modulus in the range of from 10 to 4500 MPa.

3. The polymeric molding composition according to claim 1 or 2, which comprises from 10 to 80 % by weight of soft material selected from the group of crosslinked or uncrosslinked ethylene-propylene copolymers whose ethylene content is greater than 20 % by weight, crosslinked or uncrosslinked ethylene-butene copolymers, crosslinked or uncrosslinked ethylene-hexene copolymers, crosslinked or uncrosslinked ethylene-propylene-diene-copolymer (EPDM), or else crosslinked or uncrosslinked synthetic or natural rubbers, crosslinked or uncrosslinked styrene-ethylene-butadiene terpolymers (SEBS), crosslinked or uncrosslinked polybutylene, or PVC.

4. The polymeric molding composition according to one or more of claims 1 to 3, which comprises from 5 to 50 % by weight of glass material as filler, based on the total weight of the molding composition, where the glass material is spun glass fibers or is chopped spun glass fibers whose average length is in the range of from 100 µm to 10 mm or is glass particles in the form of ground glass or milled glass fibers whose average particle size is in the range of from 3 µm to 5 mm, or glass beads or hollow glass beads whose average bead diameter is in the range of from 3 µm to 2 mm.

5. The polymeric molding composition according to one or more of claims 1 to 4, which comprises, if appropriate in combination with the soft material with the glass material and with the thermoplastic polymer also mineral fillers.

6. The polymeric molding composition according to claim 4 or 5, which comprises from 10 to 40 % by weight of chopped spun glass fibers, if appropriate in combination with other manifestations of glass material or with mineral fillers.

7. The polymeric molding composition according to one or more of claims 1 to 6, which comprises an amount of from 5 to 50 % by weight of thermoplastic polymer, based on the total weight of the molding composition, the thermoplastic polymer used comprising propylene homopolymer or polypropylene-ethylene copolymer whose ethylene content is smaller than 20 % by weight, or polyamide, polyester, polystyrene, polycarbonate, or acrylonitrile-butadiene-styrene terpolymer (ABS), or PVC or the like.

8. The polymeric molding composition according to claim 7, which comprises, as thermoplastic polymer, an amount of from 10 to 40 % by weight of propylene homo- or copolymers.

9. The polymeric molding composition according to one or more of claims 1 to 8, which also comprises a polar-functionalized compatibilizer selected from the group comprising peroxides or silanes, such as aminosilanes, epoxysilanes, amidosilanes, acrylic silanes, or maleic anhydride-grafted polyolefins.

10. The polymeric molding composition according to one or more of claims 1 to 9, which also comprises stabilizers with respect to heat, stabilizers with respect to UV radiation, or stabilizers with respect to oxidative degradation, or pigments, or antistatic agents, or flame retardants, or other additives, each of them in advantageous amounts.

11. A process for the preparation of a molding composition according to one or more of claims 1 to 10, in which the individual components are brought together and are mixed and/or kneaded at temperatures in the range of from 120 to 400 °C and are cooled subsequently.

12. The use of a polymeric molding composition according to one or more of claims 1 to 10 for the preparation of moldings for the automotive industry or for household devices.

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# INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L23/02 C08K3/40
ADD. C08L23/10 C08L23/16 C08L23/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08L C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
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C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| X<br>Y | US 5 916 953 A (JACOBY PHILIP [US] ET AL)<br>29 June 1999 (1999-06-29)<br>the whole document | 1-12<br>1-12 |
| X | GB 2 312 427 A (NISSAN MOTOR [JP]; NIHON PLAST CO LTD [JP]; CHISSO CORP [JP])<br>29 October 1997 (1997-10-29)<br>the whole document | 1-12 |
| X | GB 2 143 831 A (MITSUBISHI PETROCHEMICAL CO) 20 February 1985 (1985-02-20)<br>the whole document | 1-12 |
| Y | JP 05 154962 A (SEKISUI CHEMICAL CO LTD)<br>22 June 1993 (1993-06-22)<br>abstract | 1-12 |

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[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

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International application No
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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| Y | WO 2005/040263 A (BASELL POLYOLEFINE GMBH [DE]; MECKLENBURG THOMAS [DE]; ROHRMANN JUERGE) 6 May 2005 (2005-05-06) the whole document | 1-12 |

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2006/008268

| Patent document cited in search report | Publication date | Patent family member(s) | Publication date |
|---|---|---|---|
| US 5916953 A | 29-06-1999 | NONE | |
| GB 2312427 A | 29-10-1997 | JP 3492847 B2 | 03-02-2004 |
| | | JP 9290674 A | 11-11-1997 |
| | | US 2001008915 A1 | 19-07-2001 |
| GB 2143831 A | 20-02-1985 | JP 1007618 B | 09-02-1989 |
| | | JP 1520699 C | 29-09-1989 |
| | | JP 59226041 A | 19-12-1984 |
| JP 5154962 A | 22-06-1993 | NONE | |
| WO 2005040263 A | 06-05-2005 | EP 1675899 A1 | 05-07-2006 |