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**WO 2006/063698 A1**

(54) Title: POLYPROPYLENE COMPOSITION HAVING IMPROVED SCRATCH RESISTANCE

(57) Abstract: The invention relates to a polypropylene composition comprising: (i) 30-97 wt.% of a polypropylene; (ii) 2-20 wt.% of a branched low density polyethylene with a density between 910 and 935 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min; (iii) 1-20 wt.% of a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin with a density between 840 and 890 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min; and (iv) 0.5 -60 wt.% of a filler, wherein wt.% are calculated on the total amount of the polypropylene composition.

## POLYPROPYLENE COMPOSITION HAVING IMPROVED SCRATCH RESISTANCE

The present invention is directed to a polypropylene composition having improved scratch resistance properties.

According to Wolsske et al. in the "Effect of composition on the scratch resistance of filled TPO's" (SPE; TPO conference; year 2001; pages 225-230) the enhancing of the scratch resistance of TPO's such as for example polypropylene is still an active goal of materials design to extend their use to high visibility exterior and interior automotive parts. Wolsske teaches that fatty acid amide based lubricants are effective in enhancing the scratch resistance.

It is the object of the present invention to provide a polypropylene composition having improved scratch resistance in combination with the other desired properties.

The present invention is characterized in that the polypropylene composition comprises:

- (i) 30 –97 wt.% of a polypropylene (PP);
- (ii) 2 –20 wt.% of a long-chain branched low density polyethylene (LDPE) with a density between 910 and 935 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min;
- (iii) 1–20 wt.% of a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin with a density between 840 and 890 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min; and
- (iv) 0,5 –60 wt.% of a filler,

wherein wt.% are based on the total weight of the polypropylene composition.

The presence of the specific combination of the specific branched low density polyethylene and the specific copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin in the polypropylene composition enhances the scratch resistance.

Furthermore the polypropylene composition has an excellent combination of properties such as for example impact resistance, elongation at break, modulus and dimensional stability.

A further important advantage of the composition according to the present invention is that the composition can be injection moulded at a high injection rate while the moulded part thus obtained does not exhibit (or at least exhibits them to a strongly reduced extent) surface defects in the form of a stripe or a pattern.

The polypropylene in the composition according to the invention may be a homopolymer as well as a copolymer, particularly all kinds of reactor copolymers, for instance block copolymers (blends of a homopolymer and a rubber, for instance an EPR rubber) may be used. Many types of polypropylene are commercially available and they can be readily prepared by methods well known and extensively described in the art using for instance conventional Ziegler-Natta catalysts as well as single site catalysts, for instance metallocene catalysts. Of course, also mixtures of different types of polypropylene can be used.

5 The MFI of the polypropylene (measured in accordance with ISO 10 1133 using a 2.16 kg weight and at a temperature of 230 °C) may range between wide limits, for instance between a lower limit of 0.1 g/10min and an upper limit of 100 g/10min. Preferably the lower limit is 10 g/10min and more preferably the lower limit is 20 g/10min.

15 Generally, the polypropylene homopolymer has an isotactic structure, which means that the isotacticity is high, for instance higher than 95% and preferably higher than 98%.

20 According to another preferred embodiment of the invention the propylene is a copolymer of propylene and of an olefin other than polypropylene. The olefin other than propylene may be for instance an  $\alpha$ -olefin, in particular 1-alkene having for instance 2 or 4-20 C-atoms or cyclic olefins, optionally containing more than one ring, having a double bond in the ring structure. Examples of suitable olefins include ethylene, butene, hexene, styrene, cyclopentene and/or norbornadiene.

25 Preferably, the  $\alpha$ -olefin is a 1-alkene having 2 or 4-8 C-atoms.

More preferably, the  $\alpha$ -olefin is ethylene.

According to a further preferred embodiment of the invention the composition comprises 60- 80wt% polypropylene.

30 In order to enlarge the impact resistance the polypropylene may contain rubber. The polypropylene composition may comprise in an amount of, for instance, 0-30 wt.% of a rubber, preferably 5-20 wt.%, with a comonomer content of, for instance 10 –70 wt.%, preferably 30 –65 wt.%.

Preferably, the rubber is an ethylene propylene rubber (EPR rubber). Preferably, the EPR rubber has an ethylene content of, for instance 20 –70% and more preferably of 50 –65 wt.%.

The polypropylene may also contain a modified polypropylene.

Examples of suitable modified polypropylenes are polypropylenes grafted with for instance an unsaturated organic compound, for instance a carboxylic acid, an anhydride, an ester, or salts thereof or maleic, fumaric, (meth)acrylic, itaconic or cinnamic acid or anhydride, ester or salt thereof. Preferably maleic anhydride is used.

5 The amount of modified polypropylene may vary within wide limits. For economical reasons the amount normally will be rather low, for instance lower than 10 wt.%, preferably 0-3 wt.% modified polypropylene calculated with respect to the total amount of polypropylene.

10 The composition according to the invention contains 2-20 wt.% long-chain branched low density polyethylene (LDPE).

According to a preferred embodiment of the invention the composition contains 5-15 wt.% long-chain branched low density polyethylene.

15 Long-chain branching describes the configuration of the polymer molecule possessing side chains with a degree of polymerization of the same order as that of the main chain.

The density of the polyethylene ranges between 910 kg/m<sup>3</sup> and 935 kg/m<sup>3</sup> and the melt index ranges between 0.10 dg/minute and 100 dg/minute preferably between 8 to 80 g/10min (the MFI is measured in accordance with ISO 1133 using a 2.16 kg weight and at a temperature of 190°C).

20 Preferably, the density of the polyethylene ranges between 915 kg/m<sup>3</sup> and 935 kg/m<sup>3</sup>

Preferably, the melt index of the polyethylene ranges between 50 and 70 g/10min.

25 The composition according to the invention comprises 1-20 wt.% of a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin.

The copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin has a density between 840 and 890 kg/m<sup>3</sup>.

30 The MFI of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin is between 0.1 and 100 g/10min (measured in accordance with ISO 1133 using a 2.16 kg weight and at a temperature of 190 °C).

Preferably the density of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin is between 860 and 880 kg/m<sup>3</sup>.

Preferably the MFI of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin is between 0.1 and 10 g/10min.

The composition according to the invention may comprise a mixture of copolymers of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin.

Preferably the polypropylene composition according to the invention comprises 1-18 wt.% of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin.

5 According to a further preferred embodiment of the invention the polypropylene composition comprises 5-15 wt.% of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin.

Suitable fillers include for example talc, glass beads, glass fibers, clay, mica, wollastonite, calcium carbonate, silica, carbon black and/or titanium oxide.

10 According to a preferred embodiment of the invention the filler is talc, glass fiber or carbon black.

The amount of filler in the polypropylene composition according to the invention is between 0,5 and 60 wt.%. Fillers amounts above 40 wt.% will normally be present in mixtures of a homopolymer of polypropylene and filler or in concentrates (i.e. 15 compositions that normally will be mixed with other compositions before being further processed, for instance moulded).

Compositions used for further processing into articles, for instance by blow moulding, injection moulding, extrusion, or extrusion compression moulding, preferably contain for instance more than 1 wt.% filler, particularly more than 2 wt.% 20 filler, most preferably more than 5 wt.% filler and less than 45 wt.% filler, particularly less than 30 wt.% filler, most preferably less than 20 wt.% filler.

Preferably, the composition according to the invention comprises in addition to said components a slip agent. The amount of slip agent to be used may range between wide limits, for instance between 0 and 3 wt.%. Preferably the amount 25 of slip agent ranges between 0.1 and 1 wt.%, calculated with respect to the polypropylene composition.

Suitable slip agents include for example fatty acid amides and/or polysiloxanes.

Suitable fatty acid amides are for instance saturated and unsaturated 30 amides or saturated and unsaturated alkylenebisamides. Examples of suitable amides include palmitamide, stearyl arachideamide, stearamideoethylstearamide, stearamidoethylpalmitamide, erucamide, oleamide, erucamidoethylerucamide, stearamidoethylerucamide and/or oleamidoethylerucamide.

According to a preferred embodiment of the invention the slip agent

is a fatty acid amide.

The composition according to the invention may optionally contain additives, for instance nucleating agents and clarifiers, stabilizers, release agents, fillers, plasticizers, anti-oxidants, colorants, ignition resistant additives, lubricants, mold releases, flow enhancers and/or antistatics. These additives are well known in the art and the skilled person will select the type and amount of additives such that they do not detrimentally influence the aimed properties of the composition. The amount of additives may vary between wide limits, for instance from 0 up to 20 wt.%, particularly between 1 and 10 wt.%, calculated with respect to the polypropylene composition.

The propylene copolymer composition according to the invention is suitable to be used for instance in blow molding, injection molding or extrusion of moldings, films, sheets or pipes to be applied in a wide range of articles such as for instance, exterior and interior automotive parts, especially esthetic (non painted visible) parts, such as for example bumper beams, bumper fascia, instrument panels and pillars, thin wall packaging, caps and closures, pails and containers, crates and boxes, cans and bottles, appliance housings, houseware, toys, multi-media packaging, electrical applications, for example electrical equipment housing, lawn and garden furniture and construction sheet.

The propylene copolymer composition according to the invention is very suitable to be applied in visible interior automotive parts such as for example instrument panels and pillars.

The preparation of the compositions according to the invention may be performed according to any suitable mixing procedure. Such procedures are well known in the art such as for example dry blending the individual components, optionally followed by melt mixing, and subsequently extrusion or directly injection moulding.

The invention will be elucidated by the following examples without, however, being restricted thereto.

#### Examples

##### Example I

A composition was obtained by compounding the ingredients according to Table 1 at 210 °C in a co-rotating twin-screw extruder. The composition was then injection moulded into plates of 65 x 65 x 3.2 mm according to ISO 37-2. The appropriate samples were machined out of the plates.

The test results are given in Table 1. The amounts used are given in

wt.% calculated with respect of the total composition.

Comparative Example A

Example I was repeated except that LDPE was replaced by HDPE.

5

The composition and test results of the composition are given in

Table 1

The test result shows that the specific combination of the branched low density polyethylene and the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin in the 10 polypropylene composition enhances the scratch resistance.

Table 1

Composition		Example I	Comparative Example A
Polypropylene <sup>1</sup>	wt.%	59.3	59.3
LDPE <sup>2</sup>	wt.%	8	-
HDPE <sup>3</sup>	wt.%	-	8
Copolymer <sup>4</sup>	wt.%	10	10
Filler <sup>5</sup>	wt.%	20	20
Slip Agent <sup>6</sup>	wt.%	0.6	0.6
Additives <sup>7</sup>	wt.%	2.1	2.1
MFI (ISO 1133)	dg/min	27	25
Izod Impact (notched) @ 0 °C (ISO 180 4A)	kJ/m <sup>2</sup>	6	7
Izod Impact (notched) @ 23 °C (ISO 180 4A)	kJ/m <sup>2</sup>	34	24
E-modulus ASTM D790)	N/mm <sup>2</sup>	1600	1700
Shrink (ISO 294 3&4)	%	0.77	0.79
Scratch Resistance dL		0.5	2.1
Elongation at break (ISO R 37/2)	%	100	73

- <sup>1</sup>=SABIC Polypropylene copolymer (density ≈ 0.905 g/cm<sup>3</sup>; MFI ≈ 40 g/10min; E-modulus = 1400N/mm<sup>2</sup>);
- <sup>2</sup>=SABIC LDPE (density 0.919 g/cm<sup>3</sup>; MFI = 65 g/10min);
- <sup>3</sup>=DEX Plastomers HDPE STAMYLEX 9089

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- <sup>4</sup>= Engage® 8200 of DOW
- <sup>5</sup>=Talc Steamic® OOS D of Luzenac;
- <sup>6</sup>=oleamide (UNISLIP 1759 of CIBA)
- <sup>7</sup>= 0.1 wt.% Irganox B225 of CIBA and 2 wt.% colour master batch containing 5 40 wt.% carbon black

The scratch resistance was measured according to the VW PV 3952 scratch method with a load of 10N using VW texture K59. (VW means Volkswagen)

10 The colour of the scratch was measured relative to the unscratched surface. The colour was measured according to ISO 7724 (1984). The resulting value dL is a measure for the scratch resistance because a low dL value corresponds to a high scratch resistance.

CLAIMS

1 A polypropylene composition comprising:

5 (i) 30–97 wt.% of a polypropylene;

(ii) 2–20 wt.% of a branched low density polyethylene with a density between 910 and 935 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min;

10 (iii) 1–20 wt.% of a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin with a density between 840 and 890 kg/m<sup>3</sup> and a Melt Flow Index (MFI) between 0.1 and 100 g/10min; and

(iv) 0,5 –60 wt.% of a filler,

wherein wt.% are calculated on the total amount of the polypropylene composition.

2 A composition according to Claim 1, wherein the amount of the branched low density polyethylene is between 5 and 15 wt.%.

15 3 A composition according to any one of Claims 1-2, wherein the density of the branched low density polyethylene is between 915 and 925 kg/m<sup>3</sup>.

4 A composition according to any one of Claims 1-3, wherein the branched low density polyethylene has a Melt Flow Index between 8 and 80 g/10min.

20 5 A composition according to any one of Claims 1-4, wherein the amount of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin is between 1 and 18 wt.%.

6 A composition according to any one of Claims 1-5, wherein the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin has a density between 860 and 880 kg/m<sup>3</sup>.

7 A composition according to any one of Claims 1-6, wherein the MFI of the copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> α-olefin is between 0.1 and 10 g/10min.

25 8 A composition according to any one of Claims 1-7, wherein the composition comprises between 60 and 80 wt.% of a polypropylene.

9 A composition according to any one of Claims 1-8, wherein the composition in addition comprises a slip agent.

30 10 A composition according to any one of Claims 1-9, wherein the amount of filler is 1-45 wt.%.

11 An article made of a composition according to any one of Claims 1-10.

12 An interior automotive part made of a composition according to any one of Claims 1-10.

# INTERNATIONAL SEARCH REPORT

International application No  
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**A. CLASSIFICATION OF SUBJECT MATTER**  
INV. C08L23/10

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

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)

EPO-Internal

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 585 420 A (GRASMEDER ET AL) 17 December 1996 (1996-12-17) the whole document -----	1-12
A	US 2004/014891 A1 (KRABBENBORG FRANCISCUS J.T ET AL) 22 January 2004 (2004-01-22) the whole document -----	1-12
P, A	EP 1 591 482 A (DU PONT-MITSUI POLYCHEMICALS CO., LTD; TOHCELLO CO., LTD) 2 November 2005 (2005-11-02) the whole document -----	1-12

Further documents are listed in the continuation of Box C.

See patent family annex.

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**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International application No

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