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(54) **Title:** THERMOPLASIC VULCANIZATES COMPOSITION HAVNG IMPROVED FOAMING FORMABILITY AND MOLDED ARTICLE MANUFACTURED THEREFROM

(57) **Abstract:** The present invention relates to a thermoplastic vulcanizate composition. More specifically, the present invention relates to a thermoplastic vulcanizate composition comprising a ethylene-propylene-diene rubber, paraffin oil, a high melt-strength polypropylene having an elongational viscosity of 1.0×10^6 to 1.0×10^7 poise, an olefin block copolymer, a mineral filler, a compatibilizing agent, a crosslinker and a stabilizer. The thermoplastic vulcamzate composition of the present invention has not only a low specific gravity but also excellent mechanical properties so that it can be foamed at a high ratio and advantageously the cells in the foam are less likely to be destructed. Accordingly, it is possible to manufacture molded articles with excellent mechanical properties and a lighter weight so that it is expected to be applied in various fields regarding materials, such as weather strips for automobiles, window gaskets for buildings or the like.



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**THERMOPLASTIC VULCANIZATES COMPOSITION HAVNG IMPROVED
FOAMING FORMABILITY AND MOLDED ARTICLE MANUFACTURED
THEREFROM**

5 **[CROSS REFERENCE TO RELATED APPLICATION]**

The present application claims priority to and the benefit of Korean patent application No. 2010-0057573 filed in the Korea Intellectual Property Office on June 17, 2010, the entire content of which is incorporated hereinto by reference.

10 **[TECHNICAL FIELD]**

The present invention relates to thermoplastic vulcanizate compositions and molded articles manufactured therefrom.

[BACKGROUND OF ART]

15 Thermoplastic vulcanizates (TPV), which are a resin with thermoplastic and elastic properties, refer to a composition whose main components are crosslinked ethylene-propylene-diene rubbers (hereinafter, referred to as "EPDM") and polypropylene.

20 The crosslinked EPDM is molded into foams and used as weather seals for the automotive industry. This is because the elastomeric characteristics of the crosslinked EPDM allow it to conform to the shapes needed and to effectively hinder the entry of the noise, dust and moisture from the outside of cars.

25 However, a process for forming foamed profiles of the EPDM and its vulcanization process require a lot of installations, times and cautions, leading to declined productivity.

30 In this regard, many studies have been conducted in order to compensate the aforementioned defects and the properties of the EPDM. For example, U.S. Patent No. 7,319,121 discloses foams having smooth surfaces, a low water absorption, and improved mechanical properties, which are prepared by foaming a composition comprising EPDM, styrene-ethylene-propylene-styrene copolymers (SEPS), acrylic-modified polytetrafluoroethylenes, long-chain branched polyolefins and the like.

However, the foregoing compositions have a high specific gravity and their foaming ratio does not exceed in the order of 2 times so that there is a limit in achieving a lower density for molded articles produced therefrom. Also, at a foaming process, mechanical properties such as compression set, tensile strength, and tear strength decrease so sharply that the cells in the foam end up being easily destructed. Therefore, there remains an urgent need for developing thermoplastic vulcanizates having excellent foam formability and satisfactory mechanical properties.

[DETAILED DESCRIPTION OF THE INVENTION]

10 [TECHNICAL OBJECTIVES]

The present invention provides thermoplastic vulcanizate compositions having not only a low specific gravity but also excellent mechanical properties.

Further, the present invention provides a method of foaming the thermoplastic vulcanizate composition.

15 Further, the present invention provides molded articles manufactured from the thermoplastic vulcanizate composition.

[TECHNICAL SOLUTION]

In accordance with an embodiment of the present invention, a thermoplastic vulcanizate composition comprises:

10 to 60% by weight of an ethylene-propylene-diene rubber;

10 to 60% by weight of paraffin oil;

5 to 40% by weight of a high melt-strength polypropylene having an elongational viscosity of 1.0×10^6 to 1.0×10^7 poise;

25 5 to 40% by weight of an olefin block copolymer;

1 to 30% by weight of an organically modified mineral filler;

0.5 to 10% by weight of a compatibilizing agent;

0.05 to 5% by weight of a crosslinker; and

0.05 to 3% by weight of a stabilizer.

30 In the ethylene-propylene-diene rubber, it is preferable that the content of ethylidene norbornene (ENB) is 4.5 to 12% by weight, the content of ethylene is 50 to

80% by weight, and Mooney viscosity (ML1+8, 125 °C) is from 45 to 70.

Preferably, the paraffin oil has a kinematic viscosity of 90 to 180 cSt at 40 °C.

The weight average molecular weight of the high melt-strength polypropylene is preferably from 200,000 to 500,000.

5 The olefin block copolymers have a melt index of 0.3 to 3.0 g/10 minutes under the condition of 190 °C and 2.16kg.

The mineral filler is preferably a quaternary ammonium salt modified montmorillonite.

The mineral filler has an average particle diameter of 1 to 30µm.

10 The compatibilizing agent can be a maleic anhydride grafted polypropylene; especially wherein maleic anhydride is preferably grafted in an amount of 3 to 10% by weight.

In addition, according to other embodiments of the present invention is provided a method of foaming the thermoplastic vulcanizate composition by using an
15 extruder with a ratio between the length and the outer diameter (L/D) of at least 28 under the conditions that the process temperature is 140-210 °C and the feed rate of foaming gas is 0.40-0.80 ml/min.

In addition, according to another embodiment of the present invention are provided molded articles manufactured from the thermoplastic vulcanizate composition.

20 The molded article can be used as weather strips for automobiles, window gaskets for buildings, or the like.

[ADVANTAGEOUSEFFECT OF THE INVENTION]

The thermoplastic vulcanizate composition of the present invention has not
25 only a low specific gravity but also excellent mechanical properties so that it can be foamed at a high ratio and advantageously the cells in the foam are less likely to be destructed. Accordingly, it is possible to manufacture molded articles with excellent mechanical properties and a lighter weight so that it is expected to be applied in various fields regarding materials, such as weather strips for automobiles, window gaskets for
30 buildings or the like.

[MODE FOR PRACTICING THE INVENTION]

Hereinafter, the thermoplastic vulcanizate composition, the method of foaming the composition, and the molded articles produced from the composition will be explained.

5 During a series of studies as to thermoplastic vulcanizate compositions, the present inventors have found the following and completed the present invention based on them: mixing ethylene-propylene-diene rubbers with a high melt-strength polypropylene, an olefin block copolymer, an organically-modified mineral filler, a compatibilizing agent, and the like makes possible lowering the specific gravity of the
10 composition while foaming it at a higher ratio, and thereby it is possible to produce a molded article with a lighter weight (i.e., a lower specific gravity). Furthermore, it possesses excellent mechanical properties so that the cells in the foam are less likely to be destructed.

 In accordance with an embodiment of the present invention, a thermoplastic
15 vulcanizate composition comprises:

- 10 to 60% by weight of an ethylene-propylene-diene rubber;
- 10 to 60% by weight of paraffin oil;
- 5 to 40% by weight of a high melt-strength polypropylene having an elongational viscosity of 1.0×10^6 to 1.0×10^7 poise;
- 20 5 to 40% by weight of an olefin block copolymer;
- 1 to 30% by weight of an organically modified mineral filler;
- 0.5 to 10% by weight of a compatibilizing agent;
- 0.05 to 5% by weight of a crosslinker; and
- 0.05 to 3% by weight of a stabilizer.

25

 The ethylene-propylene-diene rubber (hereinafter, referred to as "EPDM") can be a terpolymer derived from at least two monoolefin monomers (having from 2 to 10, preferably from 2 to 4 carbon atoms) and at least one polyunsaturated olefin monomer (having from 5 to 20 carbon atoms).

30 The monoolefin monomers can be $\text{CH}_2=\text{CH-R}$ (wherein R is H or an alkyl having from 1 to 12 carbon atoms), respectively, and they can be preferably ethylene

and propylene. Preferable repeating units by at least two monoolefin monomers can be included in an amount of 90 to 99.6% by weight based on the total weight of the EPDM.

The polyunsaturated olefin monomer can be a straight chained type, a branched type, a cyclic type, a bicyclic type, a bridged ring type, or the like. Preferably, it can be a non-conjugated diene. The polyunsaturated olefin monomer can be included in an amount of 0.4 to 10% by weight based on the total weight of the EPDM.

Also, it is preferable that in the EPDM, the content of ethylidene norbornene (ENB) is at least 4.5% by weight, preferably from 4.5 to 12% by weight, and more preferably from 4.5 to 9% by weight; the content of ethylene is at least 50% by weight, preferably from 50 to 80% by weight, and more preferably from 60 to 75% by weight; and Mooney viscosity (ML 1+8, 125 °C) is at least 45, preferably 45 to 70, and more preferably 45 to 60.

In the thermoplastic vulcanizate composition according to the present invention, the EPDM can be included in an amount of 10-60% by weight, preferably 15-50% by weight, and more preferably 15-40% by weight based on the total weight of the composition.

In order to impart the minimum elasticity required for the thermoplastic vulcanizate composition, the EPDM is preferably included in an amount of at least 10% by weight based on the total weight of the composition. Also, in order to prevent deterioration of fluidity and formability due to a sharp increase in viscosity in the case of adding an excess amount of EPDM, it is preferably included in an amount of at most 60% by weight.

In this case, as the viscosity of the EPDM increases, its rubber properties such as elasticity improve while its fluidity decreases, leading to difficulties in a process for preparing an elastic body. Accordingly, the composition of the present invention includes paraffin oil as a process oil. The paraffin oil plays a role in properly decreasing the hardness of the thermoplastic vulcanizate of the present invention and increasing formability.

In order for the paraffin oil to have the aforementioned effects under the process conditions of the present invention, its kinematic viscosity at 40 °C can be at least 90 cSt, preferably from 90 to 180 cSt, and more preferably from 100 to 150 cSt.

Also, the paraffin oil can be included in an amount of 10-60% by weight, preferably 20-55% by weight, and more preferably 20-50% by weight with respect to the total weight of the composition.

In order for the addition of the paraffin oil to have an effect at a minimum, the
5 paraffin oil is preferably included in an amount of at least 10% by weight with respect to the total weight of the composition. Also, in order to prevent oozing of unabsorbed oil or a decrease in mechanical properties in the case of adding an excess amount of the paraffin oil, it is preferably included in an amount of at most 60% by weight.

If a commercial product containing paraffin oil is used for the EPDM, the total
10 content of the paraffin oil can be controlled within the aforementioned range, taking the amount of the paraffin oil contained in the commercial product of the EPDM into account.

Also, the composition of the present invention includes a high melt strength
15 polypropylene (hereinafter, referred to as "HMS-PP").

The HMS-PPs, which are a type of long-chain branched polyolefins, have excellent formability, chemical resistance, heat resistance, or the like, and are used as a component for imparting the foregoing properties together with the basic thermoplastic property in the thermoplastic vulcanizate composition of the present invention.

The melt strength of polypropylene is typically weak due to the linear structure
20 of the polymer chain and in order to deal with this, the HMS-PP can be prepared by way of lowering melt index (MI) or widening a molecular weight distribution (MWD). The most effective way to increase melt strength is introducing a long chain branch structure into backbones of polypropylene, which is generally carried out by breaking the
25 polypropylene chains and then making branch structures through secondary rearrangement.

Therefore, unlike a typical polypropylene, the HMS-PP has an increased melt strength due to the long chain branch structure and thus shows improved tensile strength, improved flexural strength, higher flexural modulus, and better scratch property. Also,
30 in a typical polypropylene, the elongational viscosity increases with an increase in time to a certain extent, after which it shows a saturated state. However, the elongational viscosity of the HMS-PP with the long chain branch structure shows a sign of increasing

even at the saturation point of the elongational viscosity in the typical polypropylene.

The HMS-PP included in the composition of the present invention has a elongational viscosity of 1.0×10^6 to 1.0×10^7 poise, and can be included in an amount of 5-40% by weight, preferably 5-30% by weight, and more preferably 5-20% by weight with respect to the total weight of the composition for the purpose of improving viscosity properties of the composition of the present invention and thus enabling foaming at a high ratio. Within the aforementioned weight range, it significantly has a synergy effect with the EPDM, improving the foaming formability and mechanical properties of the resin composition.

Also, in order to bring about such an effect, the HMS-PP has a weight average molecular weight of at least 200,000, preferably 200,000 to 500,000, and more preferably 200,000 to 350,000.

Also, the HMS-PP preferably has an elongational viscosity of 1.0×10^6 to 1.0×10^7 poise. More preferably, the high melt-strength polypropylene manufactured by Honam Petroleum Chemical Co. Ltd can be used.

Also, the composition of the present invention includes an olefin block copolymer (hereinafter, referred to as "OBC").

The OBC is a component added to impart a synergy effect of improving foam formability and mechanical properties of the resin composition together with the EPDM and the HMS-PP. In other words, the addition of the OBC brings about an increase in viscoelasticity for parts between the cells in the foam, making it possible to obtain an effect of maintaining the cells in the foam and improving mechanical properties.

The OBC contained in the composition of the present invention means a block copolymer wherein different polyolefin polymers such as ethylene, propylene, butene, isoprene, hexene, octene, or the like are linked with each other via covalent bonds. Various combinations are available such as a block copolymer consisting of a polyethylene block having a crystalline phase and a polyolefin block forming a rubber phase.

In particular, the OBC is preferably a copolymer prepared via a chain shuttling technology wherein different polymer chain blocks are alternatively generated in at least

one reactor in a tandem manner by using dual catalyst system.

In conventional random polyolefin copolymers, branched chains are grafted onto the main chains of polyethylene so that an increase in the content of comonomers leads to a decrease in T_m and heat resistance and a longer crystallization time. In contrast, even with an increase in the content of the comonomers, the OBC prepared via the chain shuttling technology has improved flexibility, improved processability, better elastic recovery, and improved compression set without causing a decrease in T_m and heat resistance, thereby making it possible to supplement deteriorated mechanical properties resulting from foaming at formation of the cells in the foam.

For having the aforementioned effects, it is preferable that the OBC has a melt index of at least 0.3 g/10min, preferably 0.3 to 3.0 g/10min, and more preferably 0.3 to 1.5 g/10min under the condition of 190 °C and 2.16kg.

Also, the OBC that has a specific gravity of at least 0.860, preferably 0.860 to 0.880, and more preferably 0.860 to 0.870 can be used. Also, the OBC that has a melting temperature (T_m) of at least 115°C, preferably 115 °C to 125 °C, and more preferably 115 to 120 °C can be used.

Also, the OBC can be included in an amount of 5-40% by weight, preferably 5-30% by weight, and more preferably 5-20% by weight with respect to the total weight of the composition. Within this weight range, it can significantly have a synergy effect of improving foam formability and mechanical properties of the resin composition together with the EPDM and the HMS-PP as described above.

Also, the composition of the present invention includes an organically modified mineral filler.

The organically modified mineral fillers are superior in compatibility and dispersibility in the composition so that adding them even in a small amount in comparison with typical mineral fillers such as kaolin and talc can have an equivalent or a better effect of addition, advantageously making it possible to lower specific gravity of the composition.

For the organically modified mineral filler, particular preference is given to a quaternary ammonium salt modified montmorillonite. More preferably, it can be the montmorillonite modified with dimethyl dihydrogenated tallow quaternary ammonium.

In addition, it is possible to use a mineral filler having an average particle diameter of 1-30 μ m, preferably 1-20 μ m, and more preferably 1-10 μ m. For the addition of the mineral filler to sufficiently have an effect of enhancing heat resistance and rigidity, its average particle diameter is preferably at least 1 μ m. In order to prevent declined workability resulting from an excessively large average particle diameter, it is preferably equal to or less than 30 μ m.

Also, in light of the effects of improving properties as described above, the specific gravity of the composition, and formation workability, the content of the mineral filler can be 1-30% by weight, preferably 1-20% by weight, and more preferably 1-10% by weight with respect to the total weight of the composition.

Also, the composition of the present invention includes a compatibilizer.

The compatibilizer is a component added for the purpose of enhancing compatibility and dispersibility of the mineral filler with the EPDM, the HMS-PP, and the OBC as described above. The mineral fillers that are dispersed non-homogeneously in the composition can cause destruction of the cells in the foam, and for the purpose of minimizing that, it is preferable to add the compatibilizer.

In particular, in the present invention, the compatibilizer is preferably a maleic anhydride grafted polypropylene (hereinafter, referred to as "PP-g-MAH"), and in this case, it is preferable that the maleic anhydride is grafted in an amount of at least 3% by weight, preferably 3-10% by weight, and more preferably 3-7% by weight.

In the composition of the present invention, using the quaternary ammonium salt modified montmorillonite as the aforementioned mineral filler and at the same time using the maleic anhydride grafted polypropylene as the compatibilizer can have a synergy effect of enhancing the mechanical properties of the cells in the foam by improving the dispersibility of the mineral fillers.

Also, the compatibilizer can be included in an amount of 0.5-10% by weight, and preferably 1-7% by weight with respect to the total weight of the composition. In order to impart an effect of improving the dispersibility of the mineral fillers, the compatibilizer should be contained in an amount of at least 0.5% by weight. Also, in order to prevent deterioration in properties of the composition due to the addition of an excessive amount of the compatibilizer, it is preferably included in an amount of 10%

by weight.

Also, the composition of the present invention includes a crosslinker.

The thermoplastic vulcanizates are prepared by adding a crosslinker for
5 crosslinking the rubber parts (i.e., the soft segments) and carrying out dynamic vulcanization with an extruder so as to have both elasticity and viscosity such as rubber.

One can select and use a crosslinker typically available in the art to which the present invention pertains, and preference is given to using a phenol resin crosslinker, a peroxide crosslinker, a silane crosslinker or the like.

10 The crosslinker can be included in an amount of 0.05-5% by weight, and preferably 0.05-3% by weight with respect to the total weight of the composition. For the purpose of preventing deterioration in properties of molded articles due to an insufficient crosslinking, the crosslinker is preferably included in an amount of at least 0.05% by weight with respect to the total weight of the composition. If the crosslinker is
15 added in an excess amount, foreign substance looking like a sunspot can be observed on the surface of the molded article and formability can deteriorate. To prevent this, the crosslinker is preferably included in an amount of at most 5% by weight.

If required, the composition of the present invention can further include a co-crosslinking agent. One can use a co-crosslinking agent typically available in the art to
20 which the present invention pertains and preferably use a metal oxide, a metal halide, or a mixture thereof. More preferably, one can use a mixture of ZnO and SnCl₂. The co-crosslinking agent can be further included in an amount of 0.05-5% by weight with respect to the total weight of the composition based on the same reason for the crosslinker.

25

Also, the composition of the present invention includes a stabilizer.

The stabilizer is added in order to avoid deterioration of the resin at the time of molding the composition, and to prevent lowering of the properties on long term use of the molded articles. One can preferably add a stabilizer typically available in the art of
30 the present invention such as an antioxidant, a heat stabilizer, a light stabilizer, a halogen scavenger or the like.

The stabilizer can be included in an amount of 0.05-3% by weight, and

preferably 0.05-1% by weight with respect to the total weight of the composition. If the addition of the stabilizer is to have the least effect, it is preferably included in an amount of at least 0.05% by weight. Also, in order to prevent occurrence of a blooming phenomenon on the surface of the molded article due to the addition of an excess amount of the stabilizer, it is preferably included in an amount of at most 3% by weight.

Also, according to other embodiment, the present invention provides a method of producing a resin from the thermoplastic vulcanizate composition and a method of foaming the resin.

10 In the method of producing a resin from the thermoplastic vulcanizate composition according to the present invention, one can use a process usually employed in the art to which the present invention pertains. Preference is given to using a reactive extrusion process.

The equipment for carrying out the reactive extrusion process includes an internal mixer and an extruder; one can use a banbury mixer or a kneader mixer for the internal mixer. For the extruder, preferable is a twin screw extruder with a ratio between the length and the outer diameter (L/D) being at least 32 and also preferable is the one equipped with at least one side feeder. The preparation process is preferably carried out at a reaction temperature of 160-220 °C.

20 Also, as a method of foaming the resin, one can use a method typically available in the art to which the present invention pertains. Preference is given to using a gas foaming method.

As the equipment for carrying out the gas foaming method, it is preferable to use an extruder equipped with a gear pump and a static mixer. A single screw extruder with a ratio between the length and the outer diameter (L/D) being at least 28 is preferable for the extruder. One can use a nucleating agent and foaming gas (CO₂, N₂, or the like) typically available in the art to which the present invention pertains. The extruder is preferably provided with at least one side feeder capable of feeding the foaming gas or the like.

30 Also, the foaming process is carried out under the conditions that a process temperature is 140-210 °C and the feed rate of the foaming gas is 0.40-0.80 ml/min.

In particular, the synergy effects resulting from inclusion of the EPDM, the

HMS, the OBC, or the like in the thermoplastic vulcanizate composition of the present invention allows the foaming ratio in the foaming process to increase up to 2.5-3.5 times, and preferably 2.8-3.3 times, advantageously making it possible to obtain a lighter weight (or a lower density) of the molded articles and at the same time providing excellent mechanical properties.

According to another embodiment of the present invention is provided a molded article manufactured from the thermoplastic vulcanizate composition.

As described above, by using the thermoplastic vulcanizate composition, the molded article can have a lighter weight (a lower density) and possesses such excellent mechanical properties that advantageously the cells in the foam are less likely to be destroyed.

Accordingly, the molded articles of the present invention are expected to be applied in various fields regarding materials such as weather strips for automobiles, window gaskets of buildings, or the like.

Hereinafter, preferred examples will be presented for a better understanding of the present invention. However, the following examples are merely illustrative of the present invention and not intended to limit the present invention.

[Manufacture of Resins]

Resins of Examples 1-2 and Comparative Examples 1-8 were prepared by mixing components at the composition ratios as set forth in Table 1. Manufacturing equipment included an internal mixer and an extruder, a twin screw extruder with a ratio between the length and the outer diameter (L/D) of 52 was used, and the process was carried out at a reaction temperature of 160-220°C.

Example 1

EPDM (manufactured by Kumho polychem, trade name: KEP-960NF, ENB content: 5.7% by weight, ethylene content: 70% by weight, Mooney viscosity (ML1+8, 125°C): 49) was added such that the content of the pure EPDM became 25.80% by weight with respect to the total composition. In the EPDM product (KEP-960NF), the content of paraffin oil was 50 PHR (Part per Hundred Resin) and the product was added such that the content of the pure EPDM became 25.80% by weight with respect to the

total composition. Also, taking the content of the paraffin oil originally contained the product into account, the content of the paraffin oil included in the total composition was controlled to be 33.55% by weight. In this case, the paraffin oil additionally added was a product from Michang Petroleum Industry (trade name: W-1900, kinematic
5 viscosity (40 °C): 137 cSt).

Also, HMS-PP (manufactured by Honam Petroleum Chemistry Co. Ltd., trade name: SMS-513, weight average molecular weight: 300,000, elongational viscosity: 1.0×10^6 to 1.0×10^7 poise) in an amount of 15% by weight with respect to the total composition;

10 OBC (manufactured by DOW, trade name: D9007, melt index (190 °C, 2.16kg): 0.5g/10min, specific gravity: 0.866, Tm: 119 °C) in an amount of 10% by weight with respect to the total composition;

as a mineral filler, montmorillonite that is modified with dimethyl dihydrogenated tallow quaternary ammonium (manufactured by Southern Clay Products,
15 trade name: Cloisite 15A (content of pure minerals: 57% by weight), average particle diameter: 6 μ m) in an amount of 8.77% by weight with respect to the total composition (content of pure minerals: about 5% by weight);

as a compatibilizer, PP-g-MAH (manufactured by Honam Petroleum Chemical Co. Ltd., trade name: PH-200, MAH graft ratio: 4% by weight) in an amount of 5% by
20 weight with respect to the total composition;

as a stabilizer, a mixture of pentaerythritol tetrakis (3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate (manufactured by Ciba, trade name: IRGANOX 1010), bis(2,4-di-tert-butylphenyl) pentaerythritol diphosphite (manufactured by: Ciba, trade name: MAGNAFLOC 6260), and a hydrotalcite-like compound (manufactured by:
25 Kyowa Chemical Industry, trade name: DHT-4A) in an amount of 0.15% by weight with respect to the total composition;

as a crosslinker, a dimethylol phenolic resin (manufactured by: SI Group, trade name: SP-1045) in an amount of 0.87% by weight with respect to the total composition;
and

30 as a co-crosslinking agent, ZnO in an amount of 0.52% by weight with respect to the total composition and SnCl₂ in an amount of 0.34% by weight with respect to the total composition were added respectively to manufacture a resin in the aforementioned

way.

Example 2

Except that each of the components was mixed at the content as set forth in Table 1, the same components were used as in Example 1. The components other than
5 the mineral filler and the PP-g-MAH were mixed and subjected to a cross-linking reaction for the EPDM before the mineral filler and the PP-g-MAH were additionally mixed therewith to prepare a resin.

Comparative Example 1

Except that a typical polypropylene (manufactured by Honam Petroleum
10 Chemical Co. Ltd., trade name: Y-120, weight average molecular weight: 200,000) was added instead of the HMS-PP, the OBC was not added and a typical type of kaolin that was not organically modified (manufactured by BASF, trade name: Satintone Whitetex Clay) was used as a mineral filler, the same components as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

Comparative Example 2

Except that a typical polypropylene (manufactured by Honam Petroleum
15 Chemical Co. Ltd., trade name: Y-120, weight average molecular weight: 200,000) was added instead of the HMS-PP, and the OBC was not added, the same components as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

Comparative Example 3

Except that the OBC was not added and a typical type of kaolin that was not
20 organically modified (manufactured by BASF, trade name: Satintone Whitetex Clay) was used as a mineral filler, the same components as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

Comparative Example 4

Except that the OBC was not added and a typical type of sodium
25 montmorillonite that was not organically modified (manufactured by Southern Clay Products, trade name: Cloisite Na⁺ (the content of pure minerals: 93% by weight), average particle diameter: 6 μ m) was added in an amount of 5.38% by weight (the
30 content of pure minerals: about 5% by weight) as a mineral filler, the same components

as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

Comparative Examples 5 to 7

Except that the OBC was not added, the same components as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

5 Comparative Example 8

Except that the OBC was not added and montmorillonite that is modified with dimethyl dihydrogenated tallow quaternary ammonium (manufactured by Southern Clay Products, trade name: Cloisite 20A (the content of pure minerals: 62% by weight), average particle diameter: 6 μ m) was added in an amount of 8.06% by weight (the
10 content of pure minerals: about 5% by weight) as a mineral filler, the same components as in Example 1 were mixed at the content as set forth in Table 1 to prepare a resin.

Table 1

Content (wt.%)		Examples		Comparative Examples							
		1	2	1	2	3	4	5	6	7	8
EPDM		25.80	28.56	29.95	30.90	29.95	32.34	33.45	32.34	30.91	31.21
Paraffin oil		33.55	37.10	38.90	40.11	38.90	42.00	43.45	42.05	40.10	40.51
PP	HMS-PP	15	12	-	-	12	13	14	13	13	13
	PP	-	-	12	13	-	-	-	-	-	-
OBC		10	10	-	-	-	-	-	-	-	-
Mine-ral filler	Cloisite 15A	8.77 (5)*	5.26 (3)*	-	8.77 (5)*	-	-	1.75 (1)*	5.26 (3)*	8.77 (5)*	-
	Cloisite 20A	-	-	-	-	-	-	-	-	-	8.06 (5)*
	Cloisite Na+	-	-	-	-	-	5.38 (5)*	-	-	-	-
	Kaolin	-	-	12	-	12	-	-	-	-	-
PP-g-MAH		5	5	5	5	5	5	5	5	5	5
Stabilizing agent		0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15
Crosslinker		0.87	0.97	1.00	1.03	1.00	1.07	1.10	1.10	1.03	1.03
Cocross-linking agent	ZnO	0.52	0.58	0.60	0.62	0.60	0.64	0.66	0.66	0.62	0.62
	SnCl ₂	0.34	0.38	0.40	0.42	0.40	0.42	0.44	0.44	0.42	0.42

(In Table 1, 0 * indicates the content of pure minerals contained in the mineral fillers)

[Measurement of the Properties of the Resin]

For the resins prepared from Examples 1 to 2 and Comparative Examples 1 to 8, properties were measured in the following way and the results were shown in Table 2.

5 (D) Specific Gravity: Measuring method ISO 1183/ Equipment as used: Electronic Densimeter

(2) Shore hardness: Measuring method ISO 868/ Equipment as used: Analogue Shore Hardness Tester

③ Tensile strength: Measuring method ISO 37(11)/ Equipment as used: 10 Universal Materials Testing Machine

Table 2

	Examples		Comparative Examples							
	1	2	1	2	3	4	5	6	7	8
Specific gravity	0.919	0.909	0.972	0.920	0.970	0.919	0.893	0.904	0.917	0.913
Shore hardness	68A	67A	68A	65A	70A	67A	63A	66A	67A	66A
Tensile strength (MPa)	7.20	7.50	6.30	5.30	6.50	4.60	4.30	5.90	6.20	4.80

As shown in Table 2, it can be found that the resins according to Examples 1 to 2 have a specific gravity lower than those of the resins of Comparative Examples 1 to 8 while having a shore hardness, and a tensile strength that are equal to or better than those of the resins of Comparative Examples 1 to 8.

In particular, it is shown that despite that the content of the mineral filler in the resins of Examples 1 and 2 are less than half of the content of the mineral filler in the resins of Comparative Examples 1 and 3, the resins of Examples 1 and 2 have not only a lower specific gravity but also excellent mechanical properties by the synergy effect resulting from the inclusion of the EPDM, the HMS-PP, the OBC, and the PP-g-MAH.

[Foaming of the Resin]

As shown in Table 3, the resins prepared as above in the examples and the comparative examples were subjected to a foaming process in the following way.

Foaming Examples 1-4

5 The resin as prepared in Example 2 was used. A single screw extruder that was provided with a gear pump and a static mixer and had a ratio between the length and the outer diameter (L/D) of 33 was used for a foaming equipment. A foaming gas (CO₂) and a nucleating agent were used and the foaming was carried out under the conditions that the process temperature was from 140 to 160 °C and the feed rate of the foaming gas
10 was from 0.50 to 0.75 ml/min.

Foaming Examples 5-7

 Except that the resin as prepared in Comparative Example 6 was used, the same equipment as in Foaming Examples 1 to 4 was utilized and the foaming was carried out under the conditions as set forth in Table 3.

15

Table 3

	Foaming Example 1	Foaming Example 2	Foaming Example 3	Foaming Example 4	Foaming Example 5	Foaming Example 6	Foaming Example 7
Types of Resins	Example 2	Example 2	Example 2	Example 2	Comp. Example 6	Comp. Example 6	Comp. Example 6
Die Temp.(°C)	140	150	150	160	140	150	160
Feed rate of gas (ml/min)	0.75	0.50	0.75	0.50	0.75	0.75	0.50
Nucleating agent (%)	-	1	1	1	-	1	1

[Measurement of Properties of the Foams]

For each of the foams prepared by Foaming Examples 1 to 7, the properties
20 were measured in the following way and the results thereof were shown in Table 4.

① Specific Gravity: Measuring method ISO 1183/ Equipment as used:

Electronic Densimeter

(2) Foaming ratio: Measuring method ISO 1183/ Equipment as used: Electronic Densimeter

5 **Table 4**

	Foaming Example 1	Foaming Example 2	Foaming Example 3	Foaming Example 4	Foaming Example 5	Foaming Example 6	Foaming Example 7
Specific Gravity	0.331	0.289	0.290	0.273	0.434	0.379	0.362
Foaming Ratio (times)	2.8	3.2	3.1	3.3	2.1	2.4	2.5

As shown in Table 4, the foams in accordance with Foaming Examples 1-4 could be foamed at a higher ratio by using the resin of Example 2 when compared with the foams of Foaming Examples 5-7. Also, in Foaming Example 5, the foaming ratio increased up to 2.1 times by using the resin of Comparative Example 6, but it was found that the resulting foam has a high specific gravity and the cells in the foam tend to be destroyed so that it has poor mechanical properties.

[CLAIMS]

1. A thermoplastic vulcanizate composition which comprises:
10 to 60% by weight of an ethylene-propylene-diene rubber;
5 10 to 60% by weight of paraffin oil;
5 to 40% by weight of a high melt-strength polypropylene having an elongational viscosity of 1.0×10^6 to 1.0×10^7 poise;
5 to 40% by weight of an olefin block copolymer;
1 to 30% by weight of an organically modified mineral filler;
10 0.5 to 10% by weight of a compatibilizing agent;
0.05 to 5% by weight of a crosslinker; and
0.05 to 3% by weight of a stabilizer.
2. The thermoplastic vulcanizate composition according to Claim 1,
15 wherein in the ethylene-propylene-diene rubber, the content of ethylidene norbornene (ENB) is from 4.5 to 12% by weight, the content of ethylene is from 50 to 80% by weight, and Mooney viscosity (ML 1+8, 125 °C) is from 45 to 70.
3. The thermoplastic vulcanizate composition according to Claim 1,
20 wherein the paraffin oil has a kinematic viscosity of 90 to 180 cSt at 40 °C.
4. The thermoplastic vulcanizate composition according to Claim 1,
wherein the weight average molecular weight of the high melt-strength polypropylene is preferably from 200,000 to 500,000.
25
5. The thermoplastic vulcanizate composition according to Claim 1,
wherein the olefin block copolymer has a melt index of 0.3 to 3.0 g/10 minutes at a condition of 190°C and 2.16kg.
- 30 6. The thermoplastic vulcanizate composition according to Claim 1,
wherein the mineral filler is a quaternary ammonium salt modified montmorillonite.

7. The thermoplastic vulcanizate composition according to Claim 1, wherein the mineral filler has an average particle diameter of 1 to 30 μ m.

8. The thermoplastic vulcanizate composition according to Claim 1,
5 wherein the compatibilizing agent is a maleic anhydride grafted polypropylene.

9. The thermoplastic vulcanizate composition according to Claim 8, wherein the compatibilizing agent is polypropylene that is grafted with the maleic anhydride in an amount of 3 to 10% by weight.

10

10. The thermoplastic vulcanizate composition according to Claim 1, further comprising 0.05 to 5% by weight of co-crosslinking agent.

11. A method of foaming a composition according to any one of Claims 1
15 to 10 by using an extruder with a ratio between the length and the outer diameter (L/D) of at least 28 at a process temperature of 140-210 °C with a feed rate of foaming gas of 0.40-0.80 ml/min.

12. A molded article manufactured from a composition according to any
20 one of Claims 1 to 10.

13. The molded article according to Claim 12, wherein it is used for a weather strip for an automobile or a window gasket of buildings.