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(54) Title: POLYETHYLENE FILM HAVING MULTI-LAYER STRUCTURE AND PACKAGING MATERIAL PRODUCED USING THE SAME

(57) Abstract: The present invention relates to a polyethylene film having a multi-layer structure and a packaging material produced using the same. Specifically, the present invention relates to a polyethylene film having a multi-layer structure that is recyclable and does not cause environmental pollution because it is formed of a polyethylene single material, has excellent processability and durability due to excellent mechanical properties and thermal properties in comparison to an existing laminated film formed of different materials, and in particular, is easily commercialized in various temperature ranges due to significantly improved durability at a low temperature, and a packaging material produced using the same.

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Description

Title of Invention: POLYETHYLENE FILM HAVING MULTI-LAYER STRUCTURE AND PACKAGING MATERIAL PRODUCED USING THE SAME

Technical Field

[1] The present invention relates to a polyethylene film having a multi-layer structure and a packaging material produced using the same.

Background Art

[2] Packaging materials have been mainly produced using a resin film formed of a resin material. Among them, a resin film formed of a polyolefin has been widely used as a packaging material because it has appropriate flexibility and excellent transparency.

[3] The resin film formed of a polyolefin has been used by bonding resin films formed of polyester, polyamide, or the like that have more excellent mechanical properties and thermal properties, and the like, with a polyurethane-based adhesive, an acrylic adhesive, or the like. Accordingly, a common packaging material includes a laminated film in which a substrate and a heat-seal layer are formed of different materials. Therefore, it is difficult to separate the different types of the resin materials of the packaging material according to the related art, and recycling is practically impossible, which has become a serious social problem in terms of the environment.

[4] In order to solve the problem, a technology for implementing a recyclable packaging material, in particular, a technology for implementing a packaging material using a resin film formed of only a polyolefin-based single material has been continuously developed. However, as described above, since mechanical properties of the polyolefin resin are inferior to those of a resin used as a substrate according to the related art, durability of a packaging material to be formed is deteriorated, such that defects are likely to occur in a printing process of the packaging material and the packaging material is easily damaged during use, such as when transported or stored. In addition, since heat resistance of the polyolefin resin is inferior to that of the resin used as a substrate according to the related art, defects are likely to occur in a heat-sealing process for forming a packaging material, such that heat-sealing properties may be deteriorated.

[5] Therefore, a film having a multi-layer structure in which polyolefin resins having mechanical and thermal properties that are different from each other based on a density, an additive, a composition ratio, and the like, in spite of using only a polyolefin-based single material, are laminated has been continuously developed. However, it is still difficult to obtain excellent mechanical and thermal properties,

commercialization is not easy in terms of cost, and in particular, there is a limit to realizing a packaging material that may be used even at a low temperature, such that improvement is required.

[6] For example, Japanese Patent Laid-Open Publication No. 2020-037189 discloses a laminated film for a packaging material in which a content of a polyolefin is 80 parts by weight or more with respect to the entire film, but there is still a problem in that durability at a low temperature is deteriorated. Japanese Patent No. 6814287 discloses a recyclable polyethylene single material laminated film, but it is still difficult to obtain excellent heat resistance and heat-sealing properties are deteriorated.

[7] Therefore, developments of a novel film that may be easily recycled to solve environmental problems because it is a resin film formed of only a single material, has mechanical and thermal properties that are not significantly deteriorated in comparison to those of a laminated film formed of different materials according to the related art, may be commercialized by maintaining excellent physical properties in various temperature ranges, and also has excellent processability, and a packaging material have been demanded.

[8] In particular, in a case where a packaging material formed of a polyethylene film having a multi-layer structure is used at a low-temperature condition, when an impact is applied due to vibration, dropping, and the like, leakage of contents to the outside frequently occurs due to cracks, tears, holes, or scratches generated in a surface area of the film. In addition, damage to the film caused by a high-temperature sealing process during heat-sealing of the film and a rupture phenomenon due to an impact applied to a heat-sealed boundary area at a low-temperature condition also frequently occur. Therefore, in order to prevent such problems, it is required for the polyethylene film having a multi-layer structure essentially to have low-temperature durability.

[9] In general, in the related art, the durability of the film is determined from a falling ball impact strength in the surface area of the film. However, the present inventors have found that in the case of the polyethylene film having a multi-layer structure, a falling ball impact strength and a puncture strength tend to be inversely proportional to each other, and have discovered that when both physical properties satisfy specific ranges of values in consideration of not only the falling ball impact strength but also the puncture strength, low-temperature durability of the film may be significantly improved, thereby completing the present invention.

[10] [Related Art Documents]

[11] [Patent Documents]

[12] (Patent Document 1) Japanese Patent Laid-Open Publication No. 2020-037189 (March 12, 2020)

[13] (Patent Document 2) Japanese Patent No. 6814287 (December 22, 2020)

Disclosure of Invention

Technical Problem

[14] An object of the present invention is to provide a polyethylene film having a multi-layer structure and a packaging material produced using the same that do not cause environmental pollution because they are recyclable and may minimize damage during processing and commercialization processes because mechanical properties and thermal properties are not significantly deteriorated in comparison to those of an existing laminated film formed of different materials and a packaging material produced using the same.

[15] Another object of the present invention is to provide a polyethylene film having a multi-layer structure that is suitably used for a molded article used in a low-temperature state, such as an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, or a refrigerated food container because a falling ball impact strength and a puncture strength are excellent in balance and thus the two mechanical properties simultaneously satisfy specific numerical ranges and durability at a low temperature is excellent, and a packaging material produced using the same.

[16] Still another object of the present invention is to provide a polyethylene film having a multi-layer structure that implements significantly excellent heat-sealing properties and high-speed bag-making processability of the film even in a low temperature range in which the film is not damaged or physical properties thereof are not deteriorated, and a packaging material produced using the same.

Solution to Problem

[17] In one general aspect, a polyethylene film having a multi-layer structure includes: an outer layer (A) containing a first ethylene polymer; an intermediate layer (B) containing a second ethylene polymer; and an inner layer (C) containing a third ethylene polymer, (A), (B), and (C) being sequentially laminated,

[18] wherein the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer satisfy the following Expressions 1 to 3, a falling ball impact strength per unit thickness and a puncture strength per unit thickness of the polyethylene film having a multi-layer structure are 3.0 to 15.0 g/μm and 0.2 to 0.4 N/μm, respectively, and a difference in melting point between the outer layer (A) and the inner layer (C) is 30 to 100°C,

[19] [Expression 1]

[20] $0.935 \leq M_1 \leq 0.968$

[21] [Expression 2]

[22] $0.900 \leq M_2 \leq 0.920$

[23] [Expression 3]

[24] $0.850 \le M_3 \le 0.905$

[25] in the Expressions 1 to 3, the M_1 is a density of the first ethylene polymer, the M_2 is a density of the second ethylene polymer, the M_3 is a density of the third ethylene polymer, the density is measured according to ASTM D-792, and a unit of the density is g/cm^3.

[26] In an exemplary embodiment of the present invention, a ratio of thicknesses of the outer layer (A), the intermediate layer (B), and the inner layer (C) may be 1:1 to 10:0.5 to 2.

[27] In an exemplary embodiment of the present invention, the melting point of the inner layer (C) may be 80°C or lower.

[28] In an exemplary embodiment of the present invention, a heat-sealing initiation temperature that is a temperature when the inner layer (C) is heat-sealed according to ASTM F2029 and a seal strength measured according to ASTM F88 is 1,000 gf or more may be 40 to 120°C.

[29] In an exemplary embodiment of the present invention, a melt index of each of the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be 0.5 to 5 g/10 min when measured at 190°C and 2.16 kg according to ASTM D 1238.

[30] In an exemplary embodiment of the present invention, a molecular weight distribution of the first ethylene polymer may be 3 to 10, and a molecular weight distribution of the second ethylene polymer and the third ethylene polymer may be 1 to 5.

[31] In an exemplary embodiment of the present invention, the falling ball impact strength per unit thickness may be 5.0 to 13.0 g/μm.

[32] In an exemplary embodiment of the present invention, the puncture strength per unit thickness may be 0.25 to 0.35 N/μm.

[33] In an exemplary embodiment of the present invention, the difference in melting point between the outer layer (A) and the inner layer (C) may be 50 to 90°C.

[34] In an exemplary embodiment of the present invention, a total thickness of the polyethylene film having a multi-layer structure may be 50 to 300 μm.

[35] In an exemplary embodiment of the present invention, the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be ethylene copolymers obtained by polymerizing ethylene and C_3-C_{18} α-olefin comonomers.

[36] In an exemplary embodiment of the present invention, the α-olefin comonomer may be one or a mixture of two or more selected from 1-propylene, 1-butene, 1-hexene, 1-heptene, and 1-octene.

[37] In an exemplary embodiment of the present invention, one or more selected from the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer

may be polymerized in the presence of a single-site catalyst.

[38] In an exemplary embodiment of the present invention, a functional layer may be laminated on the outer layer (A).

[39] In an exemplary embodiment of the present invention, the functional layer may include one or two or more selected from a barrier coating layer, a top coating layer, and a print layer.

[40] In another general aspect, there is provided a packaging material produced using the polyethylene film having a multi-layer structure.

[41] In still another general aspect, there is provided a molded article including the packaging material.

[42] In an exemplary embodiment of the present invention, the molded article may be any one selected from an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, a refrigerated food container, a shrink film, a heavy weight packaging film, an automatic packaging film, a stretch wrap, and a bag.

Advantageous Effects of Invention

[43] An aspect of the present invention is to provide a polyethylene film having a multi-layer structure and a packaging material produced using the same, and the packaging material is recyclable and does not cause environmental pollution since it is produced using a film formed of a polyethylene single material.

[44] Further, in the polyethylene film having a multi-layer structure and the packaging material produced using the same according to an aspect of the present invention, mechanical properties and thermal properties are not significantly deteriorated in comparison to those of an existing laminated film formed of different materials and a packaging material produced using the same, such that damage may be minimized during processing and commercialization processes.

[45] Further, in the polyethylene film having a multi-layer structure and the packaging material produced using the same according to an aspect of the present invention, since a falling ball impact strength and a puncture strength are excellent in balance, the two mechanical properties simultaneously satisfy specific numerical ranges and durability at a low temperature is excellent, such that the polyethylene film and the packaging material may be suitably used for a molded article used in a low-temperature state, such as an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, or a refrigerated food container.

[46] Further, the polyethylene film having a multi-layer structure according to an aspect of the present invention may have significantly excellent heat-sealing properties and high-speed bag-making processability even in a low temperature range in which the film is not damaged or physical properties thereof are not deteriorated.

Best Mode for Carrying out the Invention

[47] Hereinafter, a polyethylene film having a multi-layer structure and a packaging material produced using the same according to the present invention will be described in detail.

[48] Here, unless otherwise defined, all the technical terms and scientific terms have the same meanings as commonly understood by those skilled in the art to which the present invention pertains. The terms used in the description of the present invention are merely used to effectively describe a specific exemplary embodiment, but are not intended to limit the present invention.

[49] In addition, unless otherwise stated in the specification, a unit of an additive may be wt%.

[50] In addition, unless the context clearly indicates otherwise, the singular forms used in the specification and the scope of the appended claims are intended to include the plural forms.

[51] Hereinafter, unless otherwise particularly defined in the present specification, the term "polymer" may refer to a polymerizable compound produced by polymerizing monomers. Specifically, the polymer may include a homopolymer, a copolymer, a terpolymer, an interpolymer, and the like. The interpolymer may refer to a polymer produced by polymerizing two or more different monomers. Therefore, the generic term "interpolymer" may include a copolymer and a terpolymer. The copolymer refers to a polymer produced from two different monomers, and the terpolymer refers to a polymer produced from three different monomers.

[52] Hereinafter, unless otherwise specifically defined in the present specification, it will be understood that when an element such as a layer, a film, a thin film, a region, or a plate, is referred to as being "above" or "on" another element, it may be "directly on" another element or may have an intervening element present therebetween.

[53] An exemplary embodiment of the present invention may provide a polyethylene film having a multi-layer structure that is suitably used for a molded article used in a low-temperature state, such as an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, or a refrigerated food container because it is recyclable and does not cause environmental pollution since it is formed using a polyethylene single material, may minimize damage during processing and commercialization processes since mechanical properties and thermal properties are not significantly deteriorated in comparison to those of an existing laminated film formed of different materials and a packaging material produced using the same, in particular, has excellent durabilities in a surface area and a heat-sealed boundary area at a low temperature, and has significantly improved high-speed bag-making processability due to

excellent heat-sealing properties at a low temperature, and a packaging material produced using the same.

[54] The present invention will be described in detail as follows.

[55] A polyethylene film having a multi-layer structure according to an aspect of the present invention may include: an outer layer (A) containing a first ethylene polymer; an intermediate layer (B) containing a second ethylene polymer; and an inner layer (C) containing a third ethylene polymer, (A), (B), and (C) being sequentially laminated.

[56] In the case, the polyethylene film having a multi-layer structure according to an aspect of the present invention may simultaneously satisfy physical properties in which the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer satisfy the following Expressions 1 to 3, a falling ball impact strength per unit thickness and a puncture strength per unit thickness of the polyethylene film having a multi-layer structure are 3.0 to 15.0 g/μm and 0.2 to 0.4 N/μm, respectively, and a difference in melting point between the outer layer (A) and the inner layer (C) is 30 to 100°C.

[57] [Expression 1]

[58] $0.935 \leq M_1 \leq 0.968$

[59] [Expression 2]

[60] $0.900 \leq M_2 \leq 0.920$

[61] [Expression 3]

[62] $0.850 \leq M_3 \leq 0.905$

[63] In the Expressions 1 to 3, the M_1 is a density of the first ethylene polymer, the M_2 is a density of the second ethylene polymer, the M_3 is a density of the third ethylene polymer, the density is measured according to ASTM D-792, and a unit of the density is g/cm^3

[64] In an aspect of the present invention, more specifically, a density of the first ethylene polymer may be 0.950 to 0.968 g/cm^3 and more preferably 0.955 to 0.965 g/cm^3, a density of the second ethylene polymer may be 0.905 to 0.920 g/cm^3 and more preferably 0.910 to 0.920 g/cm^3, and a density of the third ethylene polymer may be 0.860 to 0.905 g/cm^3 and more preferably 0.860 to 0.890 g/cm^3, but the present invention is not limited thereto.

[65] In an aspect of the present invention, more specifically, the falling ball impact strength per unit thickness of the polyethylene film having a multi-layer structure may be 5.0 to 13.0 g/μm, preferably 8.0 to 13.0 g/μm, and more preferably 10.0 to 13.0 g/μm, and the puncture strength per unit thickness of the polyethylene film having a multi-layer structure may be 0.25 to 0.35 N/μm, preferably 0.25 to 0.30 N/μm, and more preferably 0.27 to 0.30 N/μm, but the present invention is not limited thereto.

[66] In this case, a falling ball impact strength refers to a value measured according to

ASTM D 1709, a puncture strength refers to a value obtained by measuring a maximum strength when the film is ruptured by a pin having a diameter of 6.5 mm that punctures the film in an inner surface direction at a speed of 500 mm/min, and the falling ball impact strength and the puncture strength per unit thickness refer to numerical values obtained by dividing the falling ball impact strength and puncture strength values measured according to the above methods by the total thickness of the film, respectively.

[67] In the cases of the polyethylene film having a multi-layer structure including the outer layer (A) containing the first ethylene polymer, the intermediate layer (B) containing the second ethylene polymer, and the inner layer (C) containing the third ethylene polymer, (A), (B), and (C) being sequentially laminated, and the packaging material produced using the same that satisfy the above physical properties, the polyethylene film and the packaging material are easily recycled because they are formed of a polyethylene single material, and mechanical properties such as impact resistance and a tensile strength are not deteriorated in comparison to those of a laminated film formed of different materials according to the related art, such that defects occurring in a printing process are unlikely to occur due to excellent printing processability, durability, and the like, and the packaging material is not easily damaged during use, such as when transported or stored, and thus may be easily commercialized.

[68] In particular, since physical properties such as durability in the surface area of the film and cracking strength in the heat-sealed boundary area in a low-temperature state are significantly excellent, the polyethylene film and the packaging material are significantly useful in various temperature ranges including a low temperature.

[69] In addition, any ethylene polymer satisfying the above physical properties may be used regardless of its type, and thus, it is possible to minimize an influence of a type of a functional group contained in the ethylene polymer, a molecular structure of the ethylene polymer, and the like, and a polyethylene film having a multi-layer structure having excellent physical properties desired in the present invention may be sufficiently obtained even when an ethylene polymer product of an inexpensive price that is easier to commercialize is used, such that versatility may be significantly improved.

[70] In particular, the polyethylene film having a multi-layer structure and the packaging material produced using the same that have a falling ball impact strength and a puncture strength per unit thickness simultaneously satisfying the above ranges may have excellent mechanical and thermal properties, and physical properties such as durability in the surface area of the film and cracking strength in the heat-sealed boundary area are significantly excellent even in a low-temperature state, such that the polyethylene film and the packaging material are significantly useful in various tem-

perature ranges including a low temperature. Therefore, the polyethylene film and the packaging material may be significantly suitably used for a molded article produced at a high temperature or room temperature and distributed and used in a low-temperature state, such as an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, or a refrigerated food container, such that their application ranges may be significantly expanded.

[71] More specifically, in order to evaluate a degree of durability in the heat-sealed boundary area at a low temperature of the packaging material produced using the polyethylene film having a multi-layer structure according to an aspect of the present invention, inner layers of the polyethylene film having a multi-layer structure are folded and fixed to face each other and heat-sealed at 0.2 MPa with a heating bar at 125°C for 1 second, a weight having a weight of 5.381 kg and a diameter of 20 mm is dropped at a speed of 4.3 km/s to a heat-sealed area of the film stored at a chamber at -20°C for 1 hour or longer, and an energy value when the heat-sealed boundary area is ruptured is measured.

[72] As a result, although not limited thereto, the measured energy value of the polyethylene film having a multi-layer structure according to an aspect of the present invention is 1.0 J or more, preferably 1.5 J or more, and more preferably 1.6 J or more, and it is confirmed that the heat-sealed boundary area is ruptured only when a significantly high energy impact is applied, which shows that durability in the heat-sealed boundary area at a low temperature is significantly excellent.

[73] In addition, it is confirmed that even when 10 ice pack samples obtained by filling 500 ml of water in packaging material samples having a width of 150 mm and a length of 240 mm and prepared from the polyethylene film having a multi-layer structure according to an aspect of the present invention and leaving the packaging material samples at -5°C for 24 hours are put in a box and the box is dropped from a height of 1 m, damage of the sample, such as cracking or tearing, does not occur at all, and even when the sample is left at room temperature for a long time after dropping, the inside water does not leak to the outside, which shows that durability of the film at a low temperature is also significantly excellent.

[74] Accordingly, the fact that in the case of the polyethylene film having a multi-layer structure and the packaging material produced using the same according to the present invention, all physical properties such as durability in the surface area of the film and cracking strength in the heat-sealed boundary area at a low temperature are significantly excellent is confirmed through a specific experiment. As described above, the polyethylene film and the packaging material may be significantly suitably used for a molded article produced at a high temperature or room temperature and distributed and used in a low-temperature state, such as an ice pack, a frozen food packaging bag,

a frozen food container, a refrigerated food packaging bag, or a refrigerated food container, such that their application ranges may be significantly expanded.

[75] In an aspect of the present invention, more specifically, the difference in melting point between the outer layer (A) and the inner layer (C) may be 50 to 90°C and preferably 55 to 90°C, but is not limited thereto.

[76] When the difference in melting point between the outer layer (A) and the inner layer (C) satisfies the above range, the polyethylene film having a multi-layer structure according to an aspect of the present invention may be subjected to a heat-sealing process even in a sufficiently low temperature range in which the outer layer (A) and the intermediate layer (B) are not damaged or physical properties thereof are not deteriorated, and even when the outer layer (A) and the intermediate layer (B) have a relatively low melting point in comparison to polyester, polyamide, or the like, which is mainly used in a laminated film formed of different materials according to the related art, the film is not damaged or physical properties thereof are not deteriorated, and excellent heat-sealing properties and high-speed bag-making processability of the film may be implemented.

[77] In particular, in a case where the high-speed bag-making processability is deteriorated, this leads to an increase in unit price of the packaging material to be produced, and thus, productivity and cost-effectiveness of the film and the packaging material are significantly deteriorated. In the polyethylene film having a multi-layer structure according to an aspect of the present invention, it is possible to improve the bag-making processability of the film and to obtain the cost-effectiveness accordingly by maintaining a large difference in melting point between the outer layer (A) and the inner layer (C).

[78] In addition, in an aspect of the present invention, the melting point of the inner layer (C) may be 80°C or lower and preferably 75°C or lower, and more specifically, may be 40 to 80°C and preferably 40 to 75°C, but is not limited thereto.

[79] In addition, in an aspect of the present invention, a heat-sealing initiation temperature for heat-sealing the inner layer (C) may be 40 to 120°C, preferably 50 to 110°C, and more preferably 50 to 100°C, but is not limited thereto.

[80] In this case, the heat-sealing initiation temperature refers to a temperature when a seal strength of the inner layer (C) is 1,000 gf or more when measured according to ASTM F2029 and ASTM F88.

[81] When the melting point and the heat-sealing initiation temperature of the inner layer (C) satisfy the above ranges, in the polyethylene film having a multi-layer structure and the packaging material produced using the same according to the present invention, heat-sealing properties and high-speed bag-making processability of the film may be more significantly implemented even in a sufficiently low temperature range in

which the outer layer (A) and the intermediate layer (B) of the film are not damaged or deterioration of physical properties thereof does not occur.

[82] In an aspect of the present invention, a ratio of thicknesses of the outer layer (A), the intermediate layer (B), and the inner layer (C) may be 1:1 to 10:0.5 to 2. In this case, the ratio of the thicknesses of the outer layer (A), the intermediate layer (B), and the inner layer (C) may be preferably 1:1 to 8:0.5 to 1 and more preferably 1:4 to 8:0.5 to 1, but is not limited thereto.

[83] When the thickness of each layer satisfies the above ratio range, the balanced and excellent falling ball impact strength and puncture strength per unit thickness may be more significantly implemented, and durability at a low temperature may be further improved.

[84] In addition, in an aspect of the present invention, a tensile strength of the polyethylene film having a multi-layer structure may be 350 kg/cm^2 or more and preferably 380 kg/cm^2 or more, but is not limited thereto. When the tensile strength satisfies the above range, the polyethylene film having a multi-layer structure according to an aspect of the present invention may have more excellent mechanical properties, and damage to the film may be further minimized in a commercialization process.

[85] In an aspect of the present invention, a melt index of each of the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be 0.5 to 5 g/10 min. In this case, the melt index of each of the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be preferably 0.5 to 3 g/10 min and more preferably 0.5 to 1 g/10 min, but is not limited thereto. In this case, the melt index refers to a value measured at 190°C and 2.16 kg according to ASTM D 1238.

[86] In addition, in an aspect of the present invention, a molecular weight distribution of the first ethylene polymer may be 3 to 10 and preferably 5 to 9, and a molecular weight distribution of the second ethylene polymer and the third ethylene polymer may be 1 to 5 and preferably 2 to 4, but are not limited thereto.

[87] When the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer satisfy the above melt index and molecular weight distribution ranges, processability and mechanical properties of the polyethylene film having a multi-layer structure according to an aspect of the present invention may be further improved.

[88] In an aspect of the present invention, a total thickness of the polyethylene film having a multi-layer structure may be 50 to 300 μm. In this case, the total thickness of the polyethylene film having a multi-layer structure may be preferably 100 to 200 μm and more preferably 120 to 150 μm, but is not limited thereto. When the total thickness satisfies the above range, durability of the polyethylene film having a multi-layer structure according to an aspect of the present invention may be further improved.

[89] In an aspect of the present invention, the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be ethylene copolymers obtained by polymerizing ethylene and C_3-C_{18} α-olefin comonomers.

[90] Specifically, the α-olefin comonomer may be one or a mixture of two or more selected from propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, and 1-octadecene, and more specifically, it is more preferable that the α-olefin comonomer is one or a mixture of two or more selected from 1-propylene, 1-butene, 1-hexene, 1-heptene, and 1-octene in terms of achieving the effects according to the present invention, but the present invention is not limited thereto. An ethylene copolymer having a high molecular weight may be produced while imparting fluidity to an ethylene homopolymer through the use of the α-olefin comonomer, and thus, the polyethylene film having a multi-layer structure containing the ethylene copolymer may have further improved mechanical physical properties such as impact resistance and a tensile strength.

[91] In an aspect of the present invention, a content of the α-olefin comonomer contained in 100 parts by weight of the ethylene copolymer may be 1 to 40 parts by weight, preferably 1 to 30 parts by weight, and more preferably 1 to 20 parts by weight, but is not limited thereto. When the content of the α-olefin comonomer satisfies the above range, rigidity and impact resistance of the ethylene polymer may be further improved, and the polyethylene film may be more easily applied alone to a molded article such as a film, injection molding, a compound, a sheet, and blow molding.

[92] In an aspect of the present invention, the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer may be obtained by appropriately selecting a polymerization method. For example, the polymerization may be performed by single polymerization by any one selected from gas phase polymerization, slurry polymerization, solution polymerization, and high pressure ion polymerization, or a multi-stage polymerization by a combination thereof using, as a polymerization catalyst, a multi-site catalyst such as a Ziegler-Natta catalyst or a single-site catalyst such as a metallocene catalyst, but this is a non-limited example, and the present invention is not limited thereto.

[93] The single-site catalyst is a catalyst capable of forming a homogeneous active species, and in general, the single-site catalyst is obtained by bringing any one compound selected from a metallocene-based transition metal compound and a non-metallocene-based transition metal compound into contact with a cocatalyst for activation.

[94] In this case, the single-site catalyst has a uniform active site structure in comparison to a multi-site catalyst, such that a polymer having a high molecular weight and high uniformity may be polymerized. Therefore, polymerization using a single-site catalyst

is preferable, and polymerization using a metallocene-based catalyst among the single-site catalysts is more preferable, but the present invention is not limited thereto.

[95] In particular, it is more preferable that one or more selected from the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer is formed by polymerization using a metallocene-based catalyst in terms of achieving the effects according to the present invention, but the present invention is not limited thereto.

[96] A method of producing a polyethylene film having a multi-layer structure according to an aspect of the present invention using the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer is not limited as long as it may achieve the effects to be obtained by the present invention. Examples thereof include an injection molding method, an extrusion molding method, an inflation method, a T-die method, a calendar method, a blow molding method, a vacuum molding method, and an air pressure molding method. It is more preferable to use an inflation method in terms of achieving the effects according to the present invention, but it is a non-limiting example, and the present invention is not limited thereto.

[97] In this case, the multi-layer structure may be implemented by one or two or more laminations and may be implemented by co-extrusion using a plurality of extruders, but the present invention is not limited thereto. In this case, in the co-extrusion, various physical properties of the film may be more significantly implemented by controlling the number of extruders.

[98] In an aspect of the present invention, a functional layer may be additionally laminated on the outer layer (A).

[99] In this case, the functional layer may include one or two or more selected from a barrier coating layer, a top coating layer, and a print layer, and more specifically, may include a print layer, an aluminum deposition layer, an oxygen barrier layer, an impact-resistant reinforcing layer, a heat-resistant reinforcing layer, and the like. Specifically, the oxygen barrier layer may be formed of ethylene vinyl alcohol (EVOH) or the like, and the impact-resistant reinforcing layer may be formed of polyamide (PA), polyester, or the like, but the present invention is not limited thereto.

[100] Another aspect of the present invention provides a packaging material produced using the polyethylene film having a multi-layer structure and a molded article including the same. In this case, examples of the molded article include an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, a refrigerated food container, a shrink film, a heavy weight packaging film, an automatic packaging film, a stretch wrap, and a bag.

[101] The packaging material according to an aspect of the present invention may maintain excellent durability and mechanical properties in a wide temperature range including a low temperature in spite of continuous temperature change in a production process at

room temperature, a heat-sealing process at a high temperature, and distribution and storage processes at a low temperature. Therefore, it is more preferable that the packaging material is applied to a molded article used at a low temperature, such as an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, or a refrigerated food container, in consideration of the effects according to the present invention, but it is a non-limiting example, and the present invention is not limited thereto.

[102] Hereinafter, the present invention will be described in more detail with reference to Examples and Comparative Examples. However, the following Examples and Comparative Examples are only examples for describing the present invention in more detail, and the present invention is not limited by the following Examples and Comparative Examples.

[103] [Physical Property Measurement Methods]

[104] (1) Density

[105] A density was measured according to ASTM D 792.

[106] (2) Melt Index

[107] A melt index was measured at 190°C and 2.16 kg according to ASTM D 1238.

[108] (3) Falling Ball Impact Strength

[109] A falling ball impact strength was measured according to ASTM D 1709.

[110] (4) Puncture Strength

[111] While a pin having a diameter of 6.5 mm punctured a film in an inner surface direction at a speed of 500 mm/min, a maximum strength when the film was ruptured was measured.

[112] (5) Tensile Strength

[113] A tensile strength was measured according to ASTM D 882. A unit of the tensile strength is kg/cm^2.

[114] (6) Heat-Sealing Properties

[115] A heat-seal strength was measured according to ASTM F2029 and ASTM F88. A heat-sealing initiation temperature was based on a seal strength of 1,000 gf or more.

[116] (7) Low-Temperature Cracking Strength in Heat-Sealed Boundary Area

[117] As an index of durability in a heat-sealed area of a packaging material at a low temperature, a total energy value required when the corresponding area was ruptured by applying an impact to the heat-sealed area of the packaging material at a low temperature was evaluated. Both inner surfaces of the film were heat-sealed at 0.2 MPa with a heating bar at 125°C for 1 second, a weight having a weight of 5.381 kg and a diameter of 20 mm was dropped at a speed of 4.3 km/s to a heat-sealed boundary area of the film stored at a chamber at -20°C for 1 hour or longer, and an energy value required when the heat-sealed boundary area was ruptured was measured.

[118] (8) Low-Temperature Packaging Drop Evaluation

[119] As an index of the durability of the packaging material at a low temperature, a degree of damage according to dropping of the packaging material at a low temperature was evaluated. 10 ice pack samples obtained by filling 500 ml of water in packaging material samples having a width of 150 mm and a length of 240 mm and prepared from the film and leaving the packaging material samples at -5°C for 24 hours were put in a box and the box was dropped from a height of 1 m, a degree of damage and whether the inside water leaked to the outside when the box was sufficiently left at room temperature were visually observed, and durability was evaluated based on evaluation criteria shown in Table 1.

[120] [Table 1]

Classification	◎ (Excellent)	O (Good)	X (Poor)
Degree of damage of sample	All 10 samples were not damaged and there was no leakage of water at room temperature	Scratches were observed in appearance, but there was no leakage of water at room temperature	Cracking or tearing was observed in one or more samples, and there was leakage of water at room temperature

[121] (9) Bag-Making Speed

[122] A bag-making speed is an index of productivity of making bags through heat-sealing processing, and represents the number of bags produced that may be reached when heat-sealing conditions are optimized.

[123] [Example 1]

[124] A first ethylene polymer [density: 0.957 g/cm^3, melting point: 131°C, melt index: 1.0 g/10 min (190°C, 2.16 kg), YUZEX 7302, SKGC] was used as a resin for an outer layer (A), a second ethylene polymer [density: 0.912 g/cm^3, melting point: 111°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Smart 121S, SKGC] was used as a resin for an intermediate layer (B), and a third ethylene polymer [density: 0.885 g/cm^3, melting point: 74°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Supreme 891, SKGC] was used as a resin for an inner layer (C). At this time, a polyethylene film having a multi-layer structure including three layers (A)/(B)/(C) was formed by co-extruding the resin by blown film molding while controlling the processing temperature for each layer from 165°C to 195°C using a total of three extruders having a screw size of 24 pi while performing filtering so that unmelted resin did not pass through the extruders. At this time, a die diameter was set to 50 mm, a die gap was set to 0.7 mm, a bubble expansion ratio at the time of production of the film was set to 2.6:1, and a height of a cooling wire cooled by air was set to 12 cm based on the die. The cooled and solidified film was

pulled by a nip roller and wound into a film roll. Thicknesses and the total thickness of the layers of the film were 30 μm/60 μm/30 μm (total 120 μm).

[125] The physical properties of the obtained polyethylene film are shown in Table 2.

[126] [Example 2]

[127] A polyethylene film having a multi-layer structure was produced in the same manner as that of Example 1, except that a first ethylene polymer [density: 0.965 g/cm^3, melting point: 133°C, melt index: 1.0 g/10 min (190°C, 2.16 kg), YUZEX 7300, SKGC] was used as a resin for an outer layer (A), a second ethylene polymer [density: 0.918 g/cm^3, melting point: 115°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Smart 181S, SKGC] was used as a resin for an intermediate layer (B), a third ethylene polymer [density: 0.885 g/cm^3, melting point: 74°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Supreme 891, SKGC] was used as a resin for an inner layer (C), and the thicknesses and the total thickness of the layers of the film were 12 μm/96 μm/12 μm (total 120 μm) in Example 1.

[128] The physical properties of the obtained polyethylene film are shown in Table 2.

[129] [Example 3]

[130] A polyethylene film having a multi-layer structure was produced in the same manner as that of Example 1, except that a first ethylene polymer [density: 0.963 g/cm^3, melting point: 132°C, melt index: 0.7 g/10 min (190°C, 2.16 kg), YUZEX 8300, SKGC] was used as a resin for an outer layer (A), a second ethylene polymer [density: 0.919 g/cm^3, melting point: 124°C, MI: 0.9 g/10 min (190°C, 2.16 kg), YUCLAIR FN810, SKGC] was used as a resin for an intermediate layer (B), a third ethylene polymer [density: 0.863 g/cm^3, melting point: 45°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Solumer 861, SKGC] was used as a resin for an inner layer (C), and the thicknesses and the total thickness of the layers of the film were 16 μm/63 μm/31 μm (total 100 μm) in Example 1.

[131] The physical properties of the obtained polyethylene film are shown in Table 2.

[132] [Comparative Example 1]

[133] A polyethylene film having a multi-layer structure was produced in the same manner as that of Example 1, except that a first ethylene polymer [density: 0.965 g/cm^3, melting point: 133°C, melt index: 1.0 g/10 min (190°C, 2.16 kg), YUZEX 7300, SKGC] was used as a resin for an outer layer (A), a second ethylene polymer [density: 0.935 g/cm^3, melting point: 126°C, MI: 1.0 g/10 min (190°C, 2.16 kg), YUCLAIR FN800M, SKGC] was used as a resin for an intermediate layer (B), a third ethylene polymer [density: 0.885 g/cm^3, melting point: 74°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Supreme 891, SKGC] was used as a resin for an inner layer (C), and the thicknesses and the total thickness of the layers of the film were 20 μm/80 μm/20 μm (total 120 μm) in Example 1.

[134] The physical properties of the obtained polyethylene film are shown in Table 2.

[135] [Comparative Example 2]

[136] A polyethylene film having a multi-layer structure was produced in the same manner as that of Example 1, except that a first ethylene polymer [density: 0.927 g/cm^3, melting point: 124°C, melt index: 0.7 g/10 min (190°C, 2.16 kg), YUCLAIR FH809, SKGC] was used as a resin for an outer layer (A), a second ethylene polymer [density: 0.902 g/cm^3, melting point: 100°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Supreme 021S, SKGC] was used as a resin for an intermediate layer (B), a third ethylene polymer [density: 0.905 g/cm^3, melting point: 102°C, MI: 1.0 g/10 min (190°C, 2.16 kg), Supreme 051S, SKGC] was used as a resin for an inner layer (C), and the thicknesses and the total thickness of the layers of the film were 30 μm/60 μm/30 μm (total 120 μm) in Example 1.

[137] The physical properties of the obtained polyethylene film are shown in Table 2.

[138] [Table 2]

	Example 1	Example 2	Example 3	Comparative Example 1	Comparative Example 2
Outer layer(A) Density/melt index (Thickness)	0.957/1.0 (30 μm)	0.965/1.0 (12 μm)	0.963/0.7 (16 μm)	0.965/1.0 (20 μm)	0.927/0.7 (30 μm)
Intermediate layer(B)Density /melt index (Thickness)	0.912/1.0 (60 μm)	0.918/1.0 (96 μm)	0.919/0.9 (63 μm)	0.935/1.0 (80 μm)	0.902/1.0 (60 μm)
Inner layer (C)Density/melt index (Thickness)	0.885/1.0 (30 μm)	0.885/1.0 (12 μm)	0.863/1.0 (31 μm)	0.885/1.0 (20 μm)	0.905/1.0 (30 μm)
Inner layer melting point (°C)	74	74	45	74	102
Difference in melting point between outer/ inner layers (°C)	57	59	87	59	22
Falling ball impact strength per unit thickness(g/μm)	6.4	10.6	13.0	1.05	15.6
Puncture strength per unit thickness(N/μm)	0.31	0.28	0.30	0.42	0.18
Tensile strength-MD (kg/cm^2)	393	424	420	351	320

Low-temperature cracking strength in heat-sealed boundary area (J)	1.67	1.64	1.87	0.97	1.95
Low-temperature packaging drop evaluation	O	◎	O	O	X
Bag-making speed	High speed (90ea/min)	High speed (90ea/min)	High speed (100ea/min)	High speed (90ea/min)	Low speed (55ea/min)

[139] Referring to Table 2, the polyethylene films having a multi-layer structure of Examples 1 to 3 satisfied all the physical properties in which the falling ball impact strength per unit thickness was 3.0 to 15.0 g/μm, the puncture strength per unit thickness was 0.2 to 0.4 N/μm, and the difference in melting point between the outer layer (A) and the inner layer (C) was 30 to 100°C. In addition, it was confirmed that the durability at a low temperature was significantly improved, such that the energy value indicating the cracking strength in the heat-sealed boundary area was 1.6 J or more, which was significantly high, significantly excellent results were exhibited in the ice pack drop evaluation, and the bag-making speed was implemented at a significantly high speed of 90 ea/min or more.

[140] On the other hand, in the cases of Comparative Examples 1 and 2 in which one or more of the outer layer (A), the intermediate layer (B), and the inner layer (C) was out of the density range according to the present invention and the falling ball impact strength per unit thickness and the puncture strength per unit thickness did not satisfy all the ranges limited by the present invention, it was confirmed that the durability at a low temperature was significantly deteriorated in comparison to those of Examples 1 to 3, and as a result of the ice pack drop evaluation, defects such as cracking or tearing occurred easily. In particular, in the case of Comparative Example 1, it was confirmed that the energy value indicating the cracking strength in the heat-sealed boundary area was 0.97 J, which was significantly low, and thus, the heat-sealed boundary area was easily ruptured.

[141] In addition, in the case of Comparative Example 2, it was confirmed that since the difference in melting point between the outer layer (A) and the inner layer (C) was 22°C, which was significantly small, the melting point of the inner layer (C) was 102°C, which was significantly high, the difference in melting point was out of the range limited by the present invention, the bag-making speed was 55 ea/min, which

was significantly low, and thus, processability and productivity in the heat-sealing process were significantly deteriorated.

[142] In addition, it was confirmed that in the case of the polyethylene film having a multi-layer structure according to the present invention, the tensile strength in a machine direction (MD) was also excellent in comparison to a laminated film formed of different materials according to the related art and Comparative Examples 1 and 2, and the polyethylene single material was used, such that the polyethylene film was easily recycled, the mechanical properties such as impact resistance were maintained or further improved, and thus, the use of the polyethylene film could be more expanded.

[143] Hereinabove, although the present invention has been described by specific matters and limited embodiments, they have been provided only for assisting in the entire understanding of the present invention. Therefore, the present invention is not limited to the above embodiments. Various modifications and changes may be made by those skilled in the art to which the present invention pertains from this description.

[144] Therefore, the spirit of the present invention should not be limited to these embodiments, but the claims and all modifications equal or equivalent to the claims are intended to fall within the scope and spirit of the present invention.

Claims

[Claim 1] A polyethylene film having a multi-layer structure, comprising:
an outer layer (A) containing a first ethylene polymer;
an intermediate layer (B) containing a second ethylene polymer; and
an inner layer (C) containing a third ethylene polymer, (A), (B), and (C) being sequentially laminated,
wherein the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer satisfy the following Expressions 1 to 3, a falling ball impact strength per unit thickness and a puncture strength per unit thickness of the polyethylene film having a multi-layer structure are 3.0 to 15.0 g/μm and 0.2 to 0.4 N/μm, respectively, and a difference in melting point between the outer layer (A) and the inner layer (C) is 30 to 100°C,
[Expression 1]
$0.935 \leq M_1 \leq 0.968$
[Expression 2]
$0.900 \leq M_2 \leq 0.920$
[Expression 3]
$0.850 \leq M_3 \leq 0.905$
In the Expressions 1 to 3, the M_1 is a density of the first ethylene polymer, the M_2 is a density of the second ethylene polymer, the M_3 is a density of the third ethylene polymer, the density is measured according to ASTM D-792, and a unit of the density is g/cm^3.

[Claim 2] The polyethylene film having a multi-layer structure of claim 1, wherein a ratio of thicknesses of the outer layer (A), the intermediate layer (B), and the inner layer (C) is 1:1 to 10:0.5 to 2.

[Claim 3] The polyethylene film having a multi-layer structure of claim 1, wherein the melting point of the inner layer (C) is 80°C or lower.

[Claim 4] The polyethylene film having a multi-layer structure of claim 1, wherein a heat-sealing initiation temperature that is a temperature when the inner layer (C) is heat-sealed according to ASTM F2029 and a seal strength measured according to ASTM F88 is 1,000 gf or more is 40 to 120°C.

[Claim 5] The polyethylene film having a multi-layer structure of claim 1, wherein a melt index of each of the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer is 0.5 to 5 g/10 min when measured at 190°C and 2.16 kg according to ASTM D 1238.

[Claim 6] The polyethylene film having a multi-layer structure of claim 1, wherein a molecular weight distribution of the first ethylene polymer is 3 to 10, and a molecular weight distribution of the second ethylene polymer and the third ethylene polymer is 1 to 5.

[Claim 7] The polyethylene film having a multi-layer structure of claim 1, wherein the falling ball impact strength per unit thickness is 5.0 to 13.0 g/μm.

[Claim 8] The polyethylene film having a multi-layer structure of claim 1, wherein the puncture strength per unit thickness is 0.25 to 0.35 N/μm.

[Claim 9] The polyethylene film having a multi-layer structure of claim 1, wherein the difference in melting point between the outer layer (A) and the inner layer (C) is 50 to 90°C.

[Claim 10] The polyethylene film having a multi-layer structure of claim 1, wherein a total thickness of the film is 50 to 300 μm.

[Claim 11] The polyethylene film having a multi-layer structure of claim 1, wherein the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer are ethylene copolymers obtained by polymerizing ethylene and C_3-C_{18} α-olefin comonomers.

[Claim 12] The polyethylene film having a multi-layer structure of claim 11, wherein the α-olefin comonomer is one or a mixture of two or more selected from 1-propylene, 1-butene, 1-hexene, 1-heptene, and 1-octene.

[Claim 13] The polyethylene film having a multi-layer structure of claim 1, wherein any one of the first ethylene polymer, the second ethylene polymer, and the third ethylene polymer is polymerized in the presence of a single-site catalyst.

[Claim 14] The polyethylene film having a multi-layer structure of claim 1, wherein a functional layer is laminated on the outer layer (A).

[Claim 15] The polyethylene film having a multi-layer structure of claim 14, wherein the functional layer includes one or two or more selected from a barrier coating layer, a top coating layer, and a print layer.

[Claim 16] A packaging material produced using the polyethylene film having a multi-layer structure of any one of claims 1 to 15.

[Claim 17] A molded article comprising the packaging material of claim 16.

[Claim 18] The molded article of claim 17, wherein the molded article is any one selected from an ice pack, a frozen food packaging bag, a frozen food container, a refrigerated food packaging bag, a refrigerated food container, a shrink film, a heavy weight packaging film, an automatic packaging film, a stretch wrap, and a bag.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR2022/007978

A. CLASSIFICATION OF SUBJECT MATTER

B32B 27/32(2006.01)i; **B32B 27/08**(2006.01)i; **B32B 7/022**(2019.01)i; **C08L 23/08**(2006.01)i

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)

B32B 27/32(2006.01); A01G 9/14(2006.01); B29C 48/08(2019.01); B29C 48/18(2019.01); B29C 65/02(2006.01); B32B 27/08(2006.01); B65D 30/02(2006.01); B65D 65/40(2006.01)

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

Korean utility models and applications for utility models
Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS(KIPO internal) & Keywords: polyethylene, laminate, density, melting point, thickness

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 2005-144757 A (FUKUSUKE KOGYO CO., LTD.) 09 June 2005 (2005-06-09) claims 1-7; paragraphs [0015]-[0044]; example 1; table 2	1-18
Y	KR 10-2261130 B1 (ANYCHEM INC.) 07 June 2021 (2021-06-07) paragraphs [0017]-[0050]; example 2; tables 2-6	1-18
Y	KR 10-2157101 B1 (GEOSUNG P&P CO., LTD.) 17 September 2020 (2020-09-17) claims 1-5; paragraphs [0036]-[0067]; example 2; table 1; figures 1-3	1-18
A	JP 2020-526412 A (DOW GLOBAL TECHNOLOGIES LLC) 31 August 2020 (2020-08-31) the whole document	1-18
A	JP 2003-251697 A (SUMITOMO CHEM CO., LTD. et al.) 09 September 2003 (2003-09-09) the whole document	1-18

☐ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

* Special categories of cited documents:
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"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
05 September 2022	**05 September 2022**

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INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/KR2022/007978

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