Title: AN ADHESIVE COMPOUND COMPOSITION, LAMINATED FILM USING IT AND USES THEREOF

Abstract: The present invention relates to an adhesive compound composition having an appropriate degree of tackiness suitable for a packaging material that can be opened easily and sealed again, a laminated film using such adhesive compound composition for the intermediate layer, and a packaging material comprising said laminated film that can be opened easily and sealed again. The present invention provides an adhesive compound composition comprising an olefin-based block copolymer elastomer (A), an ethylene-vinyl acetate copolymer having a vinyl acetate content of 25 to 80 wt% (B) and a tackifier (C), a laminated film formed by laminating a laminating layer made of a thermoplastic resin (D) and a heat-sealing layer made of a thermoplastic resin (E) via an intermediate layer made of a said adhesive compound composition, and a container enclosure material comprising said laminated film.

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DESCRIPTION

An adhesive compound composition, laminated film using it and uses thereof

Field of Technology:
The present invention relates to an adhesive compound composition having an appropriate degree of tackiness suitable for a packaging material that can be opened easily and sealed again, a laminated film using such adhesive compound composition for the intermediate layer, and a packaging material comprising such laminated film that can be opened easily and sealed again.

Packages sealing a plastic container in the form of a bottle, cup or tray with an enclosure material consisting of plastic laminated film or aluminum foil laminated film are used widely for packages of snacks such as rice crackers and potato chips and food such as jelly, milk, yogurt, pudding, soybean curd and lactic acid beverage, blister packages and other materials for packaging pharmaceuticals, medical equipment, articles of daily use, general merchandise. The physical properties required of these packaging materials are mechanical strength that will stand handling in distribution channels, sealing strength that allows hygienic conditions to be kept, and ease of opening at the time of use.

However, most of such conventional packaging materials are such that although they have superior easy-to-open properties, it was completely impossible to resealing packages again once they are opened to take out the contents of the packages.

On the other hand, various methods using an adhesive compound layer as the intermediate layer have been proposed for resealable packaging materials. Examples of such methods include containers (Patent Reference 1, for example) that can be opened and closed freely because they use an enclosure material comprising the face substrate and the back substrate bonded by means of an adhesive compound layer, which will be exposed at the time of opening, resealable packaging materials (Patent Reference 2 and 3, for example) using as the adhesive compound layer an adhesive compound comprising an elastomer such as styrene-butadiene copolymer and ethylene-vinyl acetate copolymer and a tackifier such as terpene resin and petroleum hydrocarbon resin, and multi-layer film (Patent References 4 and 5, for example) using a tackifier using hydrogenated styrene-butadiene copolymer as elastomer.


However, it has been found that even if a laminated film as mentioned in the above Patent Reference using as an intermediate layer an adhesive compound composition consisting of elastomer such as styrene-butadiene copolymer and a
tackifier such as terpene resin is used for a packaging material with easy-to-open properties, the easy-to-open properties and sealing strength of such packaging material are not balanced in some cases because heat-sealing strength (initial opening strength) is too high or resealing strength is inadequate in some cases due to an occurrence of delamination between the face resin layer and the adhesive resin layer or between the heat-sealing resin layer and the adhesive resin layer or the inadequate adhesion of the adhesive resin layer.

Disclosure of the Invention:
Issue to be resolved by the invention:
The purpose of the present invention is to obtain a packaging material that has heat sealability, easy-to-open properties, repeated resealability and excellent resealing strength and shows a good balance between easy-to-open properties and resealability.
Means for resolving the issue:
The present invention provides an adhesive compound composition comprising an olefin-based block copolymer elastomer (A), an ethylene-vinyl acetate copolymer (B) having a vinyl acetate content of 25 to 80 wt% and a tackifier (C), a laminated film comprising a heat-sealing layer made of a thermoplastic resin (E) which is laminated onto one side of a laminating layer made of a thermoplastic resin (D) via an intermediate layer made of said adhesive compound composition, a container enclosure material comprising a laminated film, a package using a laminated film, and a container using a container enclosure.
Best embodiment of the invention:
Below is given an explanation of the elements of the present invention.
Olefin-based block copolymer elastomer (A):
The olefin-based block copolymer elastomer (A) which is related to the present invention is a block copolymer that has a so-called soft segment comprising at least one olefin-based elastomer polymer in its molecule and at least one so-called hard segment. The soft segment comprising an olefin-based elastomer polymer is normally random polymers of \( \alpha \)-olefins such as ethylene, propylene and 1-butene, polymers of diene compounds (diolefins) such as butadiene and isoprene, or hydrogenated materials thereof. The hard segment is normally polymers having a glass transition temperature of not lower than normal temperature such as polystyrene polymer, those crystalline \( \alpha \)-olefin polymers of ethylene, propylene, 1-butene, etc., or those crystalline hydrogenated materials of butadiene polymer.
Preferable examples of said olefin-based block copolymer elastomer (A) include a styrene-diene block copolymer (A1) which has a soft segment comprising at least one diene block (diene polymer, diolefin polymer) and a hard segment comprising at least one crystalline olefin block (crystalline olefin polymer). These block copolymers are generally manufactured and sold as thermoplastic elastomer.
Styrene-diene block copolymer (A1):

The styrene-diene block copolymer (A1) which is related to the present invention is a block copolymer which has a soft segment comprising a diene block and a hard segment comprising a styrene block (styrene polymer). It is a type of block copolymer that is commercially manufactured and sold as a thermoplastic elastomer. Specific examples of styrene-diene block copolymer (A1) includes styrene-butadiene block copolymer (SB), styrene-butadiene-styrene block copolymer (SBS), styrene-isoprene block copolymer (SI) and styrene-isoprene-styrene block copolymer (SIS) or hydrogenated materials of these block copolymers. Such hydrogenated materials may be either block copolymers all of the styrene and diene blocks of which have been hydrogenated or block copolymers only diene block of which has been hydrogenated or partially hydrogenated materials of block copolymers parts of the styrene and diene blocks of which have been hydrogenated.

If these block copolymers or their hydrogenated materials are used as the adhesive compound composition, they may be used singly or in combination of two or more of them.

Out of these block copolymers, hydrogenated materials of the block copolymers containing a styrene block and a diene block, such as hydrogenated materials of styrene-butadiene block copolymer (SB), hydrogenated materials of styrene-butadiene-styrene block copolymer (SBS), hydrogenated materials of styrene-isoprene-styrene block copolymer (SIS) and hydrogenated materials of styrene-isoprene block copolymer (SI) are preferable. Specifically, hydrogenated materials (SEB) of styrene-butadiene block copolymer (SB), hydrogenated materials (SEBS) of styrene-butadiene-styrene block copolymer (SBS) and hydrogenated materials (styrene-ethylene/propylene-styrene block copolymer, SEPS) of styrene-isoprene-styrene block copolymer (SIS) are most preferable in that they show excellent heat stability in extrusion processing and stability in processing and control the occurrence of deteriorated materials and odor.

The styrene-diene block copolymer (A1) relating to the present invention has MFR (melt flow rate as defined in ASTM D-1238 under a load of 5 kg at the temperature of 200°C) which is not particularly specified but normally ranges from 0.5 to 20 g/10 minutes, preferably 1 to 15 g/10 minutes, so long as the adhesive compound composition formed by mixing it with ethylene-vinyl acetate copolymer (B) and tackifier (C) as described below can be formed into film.

Olefin-noncrystalline olefin block copolymer (A2):

The olefin-noncrystalline olefin block copolymer (A2) relating to the present invention is a block copolymer which has a soft segment comprising at least a noncrystalline olefin block (noncrystalline olefin polymer) in its molecule and a hard segment comprising a crystalline olefin block (crystalline olefin polymer). It is a type of block copolymer that is commercially manufactured and sold as what is called a thermoplastic elastomer.

The structure of the olefin-noncrystalline olefin block copolymer (A2) is not
particularly limited so long as its molecule has at least one each of the noncrystalline olefin block A and the crystalline olefin block B. Examples of the structure include \((B-A)n_1, (B-A)n_2 - B\) and \((A-B)n_3 - A\), where \(n_1\), \(n_2\) and \(n_3\) are whole numbers not less than 1) and structures coupling these by use of a coupling agent. Out of these, copolymers having tri-blocks and copolymers having a larger number of blocks than that are preferable.

Specific examples of the block copolymer (A2) include block copolymers which have a soft segment which comprises mainly a polymer block mainly made of a diene compound or a noncrystalline olefin block having a structure similar to that of ethylene-butene copolymer or ethylene-propylene copolymer which is obtained by hydrogenating such polymer block and a hard segment which can be made a crystalline polyolefin by hydrogenating a diene compound as the primary component. These specific examples also include a block copolymer which is obtained by replacing part of the polymer that can be made a crystalline polyolefin by hydrogenating a diene compound as the primary component with a hard segment comprising a polymer of vinyl aromatic compound such as styrene. Specific examples of such diene compound include 1,3-butadiene, isoprene, 2,3-dimethyl-1,3-butadiene, 1,3-pentadiene and 2-methyl-1,3-pentadiene.

Such polymer that can be made a crystalline polyolefin by hydrogenating a diene compound as the primary component can provide a crystalline olefin block having a structure similar to that of ethylene polymer, for example, by hydrogenating a polymer block having not more than 25% of vinyl bonds (the amount of 1,2- and 3,4-bonds contained) of polybutadiene.

Specific examples of such block copolymer (A2) include crystalline olefin (crystalline ethylene polymer)-ethyene \(\cdot\) butene (noncrystalline olefin polymer)-crystalline olefin (crystalline ethylene polymer) block copolymer (CEBC), and styrene-ethylene \(\cdot\) butene (noncrystalline olefin polymer)-crystalline olefin (crystalline ethylene polymer) block copolymer (SEBC). Various grades of these copolymers are commercially available from JSR under the trade names of DYNARON, including DR6100P, DR6200P (CEBC), DR4600P, and DR4630P (SEBC).

Other examples of such block copolymer (A2) include a propylene-propylene \(\cdot\) \(\alpha\)-olefin-propylene block copolymer which is formed by obtaining a crystalline olefin polymer by living-polymerizing a crystalline propylene-based polymer made primarily of homopolymer, or propylene and a small amount of ethylene or other \(\alpha\)-olefin such as 1-butene and then by obtaining noncrystalline olefin polymer by copolymerizing propylene and other \(\alpha\)-olefin such as ethylene and 1-butene.

Ethylene-vinyl acetate copolymer (B):

Ethylene-vinyl acetate copolymer (B) of the present invention is a copolymer of vinyl acetate with ethylene which has the vinyl acetate content in a range of 25 to 80 wt\%, preferably 30 to 80 wt\% more preferably 35 to 70 wt\%. There is a possibility that if the ethylene-vinyl acetate copolymer having a vinyl acetate
content of less than 25 wt% is mixed with the aforesaid olefin-based block copolymer elastomer (A) and tackifier (C) mentioned below and used in the intermediate layer of laminated film as an adhesive compound composition, satisfactory resealing strength will not be obtained. On the other hand, there is a possibility that if the ethylene-vinyl acetate copolymer having a vinyl acetate content of more than 80 wt% is mixed with the aforesaid olefin-based block copolymer elastomer (A) and tackifier (C) mentioned below and used in the intermediate layer of laminated film as an adhesive compound composition, satisfactory resealing strength will not be obtained.

Ethylene-vinyl acetate copolymer (B) of the present invention has MFR (melt flow rate as defined in ASTM D-1238 under a load of 2160 g at the temperature of 190°C) which is not particularly specified but normally ranges from 0.5 to 20 g/10 minutes, preferably 1 to 15 g/10 minutes, so long as the adhesive compound composition comprising mixing it with the aforesaid olefin-based block copolymer elastomer (A) and tackifier (C) mentioned below can be formed into film. Selection should be made appropriately from among the above conditions, with extrusion lamination and coextrusion properties taken into consideration.

**Tackifier (C):**

Tackifier (C) of the present invention is generally a resin-like material that is commercially manufactured and sold as a tackifier. Specific examples of tackifier (C) include coumarone resins such as coumarone indene resin; phenolic resins such as phenol formaldehyde resin and xylene formaldehyde resin; terpene-based resins such as terpene phenol resin, terpene resin (α, β-pinene resin), aromatic modified terpene resin and terpene hydride resin; petroleum hydrocarbon resins such as synthetic polyterpene resin, aromatic hydrocarbon resin, aliphatic hydrocarbon resin, aliphatic cyclic hydrocarbon resin, aliphatic/aromatic petroleum resin, aliphatic/aromatic petroleum resin, unsaturated hydrocarbon polymer, hydrogenated hydrocarbon resin and hydrocarbon-based tackifying resin; and rosin derivatives such as hydrogenated rosin ester, special rosin ester and rosin-based tackifier.

Out of these, those hydrogenated resins, hydrogenated aliphatic cyclic hydrocarbon resins, hydrogenated aliphatic alicyclic petroleum resins, hydrogenated terpene resin, hydrogenated synthetic polyterpene resins and other hydrogenated resins, pentaeerythritol ester of rosin, glycerin ester of rosin and hydrogenated rosin ester which has a softening point of not lower than 70°C, preferable in a range of 70°C to 130°C are preferable because they are well compatible with olefin-based block copolymer elastomer (A) and ethylene-vinyl acetate copolymer (B). Those having a softening point lower than 70°C show satisfactory tack strength but low adhesion and cohesive strength, while those having a softening point higher than 130°C exhibit satisfactory adhesion and cohesive strength but low tack strength.

**Adhesive compound composition:**

The adhesive compound composition is a composition comprising the
aforesaid olefin-based block copolymer elastomer (A), the aforesaid ethylene-vinyl acetate copolymer (B) and the aforesaid tackifier (C). Olefin-based block copolymer elastomer (A) is in a range of preferably 10 to 80 wt%, more preferably 20 to 70 wt%; ethylene-vinyl acetate copolymer (B), in a range of preferably 10 to 80 wt%, more preferably 20 to 70 wt%; and tackifier (C), in a range of preferably 10 to 40 wt%, more preferably 10 to 30 wt%. (However, (A) + (B) + (C) shall be 100 wt%.)

Any adhesive compound composition that lacks any of olefin-based block copolymer elastomer (A), ethylene-vinyl acetate copolymer (B) and tackifier (C) may not show a good balance between easy-to-open properties and sealing strength, exhibiting too high sealing strength (initial opening strength) or too low sealing strength, when it is used as the intermediate layer between thermoplastic resin (D) and thermoplastic resin (E).

Any adhesive compound composition that is so designed that it will contain olefin-based block copolymer elastomer (A), ethylene-vinyl acetate copolymer (B) and tackifier (C) in the above respective ranges will provide a laminated film which will better balanced between heat sealing strength (initial opening strength) and resealing strength and have easy-to-open properties and sealability.

Heat stabilizers, weathering stabilizers, ultraviolet light absorbers, lubricants, slip agents, nucleating agents, anti-blocking agents, antistatic agents, antifogging agents, pigments, inorganic or organic fillers and other various additives that are commonly added to thermoplastic resins may be added to the adhesive compound composition of the present invention or any of olefin-based block copolymer elastomer (A), ethylene-vinyl acetate copolymer (B) and tackifier (C), or both of them to such extent that will not frustrate the purpose of the present invention.

Thermoplastic resin (D):

Thermoplastic resin (D) of the present invention is any of the various thermoplastic resins known to the public. Specific examples of thermoplastic resin (D) include polyolefins (polyethylene, polypropylene, polybutene, poly-4-methyl-1-pentene, modified polyolefins modified with unsaturated carboxylic acid or its derivatives, etc.), polyesters (polyethylene terephthalate, polybutylene terephthalate, polyethylene naphthalate, glycol-modified polyethylene terephthalate, glycol-modified polybutylene terephthalate, etc.), polyamides (nylon-6, nylon-66, nylon-MXD6, etc.), polyvinyl chloride, polyimide, ethylene-vinyl alcohol copolymer, polyvinyl alcohol, polyacrylonitrile, polystyrene, acrylonitrile-styrene copolymer, acrylonitrile-butadiene-styrene copolymer, polycarbonate, ethylene-vinyl acetate copolymer (EVA), ethylene-(meth)acrylic acid copolymer or its ionomer, ethylene-(meth)acrylic acid ester copolymer and other ethylene-acid (derivative) copolymer, or their mixtures.

Examples of polyolefins include ethylene-based polymers (polyethylene) composed primarily of ethylene such as homopolymers of ethylene or random copolymers of ethylene and other α-olefins, including high-pressure-process
low-density polyethylene (HP-LDPE), linear low-density polyethylene (LLDPE), medium-density polyethylene (MDPE) and high-density polyethylene (HDPE); propylene-based polymers (polypropylene) composed primarily of propylene such as homopolymers of propylene or copolymers of propylene and other α-olefins, including polypropylene (PP), propylene-ethylene random copolymer, propylene-ethylene-1-butene random copolymer, propylene-1-butene random copolymer, and propylene-ethylene block copolymer; 1-butene-based polymers (polybutene) composed primarily of 1-butene such as homopolymers of 1-butene or copolymers of 1-butene and other α-olefins, including poly-1-butene, 1-butene-propylene random copolymer and 1-butene-ethylene random copolymer; and 4-methyl-1-pentene-based polymers (poly-4-methyl-1-pentene) composed primarily of 4-methyl-1-pentene such as homopolymers of 4-methyl-1-pentene or copolymers of 4-methyl-1-pentene and other α-olefins, including poly-4-methyl-1-pentene and 4-methyl-1-pentene-α-olefin random copolymer.

Such polyolefins may be those polyolefins the whole or any part of which is graft-modified with unsaturated carboxylic acid such as maleic acid or derivatives of acid anhydride such as maleic anhydride. Furthermore, out of these thermoplastic resins (D), ethylene-based polymers such as MDPE and HDPE, propylene-based polymers, 4-methyl-1-pentene-based polymers, polyester, polynamide, polystyrene, etc., which have a relatively high melting point and stiffness, are preferable, depending on a combination with thermoplastic resin (E) as described below.

Heat stabilizers, weathering stabilizers, ultraviolet light absorbers, lubricants, slip agents, nucleating agents, anti-blocking agents, antistatic agents, antifogging agents, pigments, inorganic or organic fillers and other various additives that are commonly added to thermoplastic resins may be added to thermoplastic resin (D) of the present invention to such extent that will not frustrate the purpose of the present invention.

Thermoplastic resin (E):

Thermoplastic resin (D) of the present invention is any of the various thermoplastic resins known to the public like the aforesaid thermoplastic resin (D). Specific examples of thermoplastic resin (D) include polyolefins (polyethylene, polypropylene, polybutene, poly-4-methyl-1-pentene, modified polyolefins modified with unsaturated carboxylic acid or its derivatives, etc.), polyesters (polyethylene terephthalate, polybutylene terephthalate, polyethylene naphthalate, glycol-modified polyethylene terephthalate, glycol-modified polybutylene terephthalate, etc.), polyamides (nylon-6, nylon-66, nylon-MXD6, etc.), polyvinyl chloride, polyamide, ethylene-vinyl alcohol copolymer, polyvinyl alcohol, polyacrylonitrile, polystyrene, acrylonitrile-styrene copolymer, acrylonitrile-butadiene-styrene copolymer, polycarbonate, ethylene-vinyl acetate copolymer (EVA), ethylene-(meth)acrylic acid copolymer or its ionomer, ethylene-(meth)acrylic acid ester copolymer and other ethylene-acid (derivative) copolymer, or their mixtures.
Examples of polyolefins include the aforementioned ethylene-based polymers, propylene-based polymers, and 1-butene-based polymers.

Out of these thermoplastic resins (E), thermoplastic resins having excellent heat sealability, including ethylene homopolymers or ethylene random copolymers with other $\alpha$-olefins whose melting point (Tm) is in a range of 90°C to 130°C, such as ultra-low density polyethylene (VLDPE), high-pressure-process low density polyethylene (HP-LDPE), linear low density polyethylene (LLDPE) and medium density polyethylene (MDPE); random copolymers of propylene and other $\alpha$-olefins whose melting point (Tm) is in a range of 110°C to 150°C, such as propylene-ethylene random copolymer, propylene-ethylene-1-butene random copolymers and propylene-1-butene random copolymer; 1-butene random copolymers whose melting point (Tm) is in a range of 75°C to 125°C such as 1-butene-propylene random copolymer and 1-butene-ethene random copolymer; ethylene-acid (derivatives) copolymers such as ethylene-vinyl acetate copolymers (EVA), ethylene-(meth)acrylic acid copolymer, ethylene-(meth)acrylic acid ester copolymer and ionomers; low-crystallinity polyesters whose melting point (Tm) is in a range of 70°C to 180°C, and styrene-based polymers such as acrylonitrile-styrene copolymer and acrylonitrile-butadiene-styrene copolymer are preferable.

Those copolymers which are called elastomers such as low-crystallinity or amorphous ethylene-$\alpha$-olefin random copolymers and propylene-$\alpha$-olefin random copolymers may be added to these thermoplastic resins (E) to improve their low-temperature heat sealability and easy-to-open properties.

Heat stabilizers, weathering stabilizers, ultraviolet light absorbers, lubricants, slip agents, nucleating agents, anti-blocking agents, antistatic agents, antifogging agents, pigments, inorganic or organic fillers and other various additives that are commonly added to thermoplastic resins may be added to thermoplastic resin (E) of the present invention to such extent that will not frustrate the purpose of the present invention.

**Laminated film:**

The laminated film of the present invention is a laminated film obtained by laminated a heat-sealing layer (heat-fusion layer) made of the aforesaid thermoplastic resin (E) onto one side of the laminating layer made of the aforesaid thermoplastic resin (D) via an intermediate layer made of the aforesaid adhesive compound composition. The laminating layer and the heat-sealing layer may be a single layer or a two-or-more-ply layer. Furthermore, the form of the laminated film of the present invention is not limited to film in form but may be tape.

The thickness of the laminated film of the present invention is determined depending on uses, but the thickness of the laminating layer is normally 1 to 100 $\mu$m, preferably 5 to 60 $\mu$m. The thickness of the intermediate layer made of the adhesive compound composition is normally 1 to 498 $\mu$m, preferably 1 to 94 $\mu$m, and the thickness of the heat-sealing layer is 1 to 20 $\mu$m, preferably 1 to 10 $\mu$m.
The surface of the laminating layer of the laminated film of the present invention may be given corona treatment, flame treatment, plasma treatment, undercoat treatment and other treatment so that its printability, adhesion with other film, slip properties, etc. will be improved.

Substrate layer:

The substrate layer of the present invention comprises a sheet-like or film-like material made of thermoplastic resin, paper, aluminum foil, etc. The thermoplastic resin is any of the various thermoplastic resins known to the public like the aforesaid thermoplastic resin (D) or the aforesaid thermoplastic resin (E). Specific examples of thermoplastic resin (D) include polyolefins (polyethylene, polypropylene, polybutene, poly-4-methyl-1-pentene, modified polyolefins modified with unsaturated carboxylic acid or its derivatives, etc.), polyesters (polyethylene terephthalate, polybutylene terephthalate, polyethylene naphthalate, glycol-modified polyethylene terephthalate, glycol-modified polybutylene terephthalate, etc.), polyamides (nylon-6, nylon-66, nylon-MXD6, etc.), polyvinyl chloride, polyimide, ethylene-vinyl alcohol copolymer, polyvinyl alcohol, polycyronitrile, polystyrene, acrylonitrile-styrene copolymer, acrylonitrile-butadiene-styrene copolymer, polycarbonate, ethylene-vinyl acetate copolymer (EVA), ethylene-(meth)acrylic acid copolymer or its ionomer, ethylene-(meth)acrylic acid ester copolymer and other ethylene-acid (derivative) copolymer, or their mixtures.

Out of these, those thermoplastic resins which have satisfactory drawability and transparency, such as polypropylene, polyethylene terephthalate and polyamide, are preferable. Furthermore, the substrate layer comprising such thermoplastic resin film may be either non-oriented film or oriented film, or extruded, extrusion-laminated or dry-laminated laminates of not less than one type of material.

Out of these substrate layers, biaxially-oriented film made of polypropylene, polyethylene terephthalate, polyamide, etc. is preferable because of its excellent transparency and stiffness.

Metals such as aluminum and zinc and inorganic oxides such as aluminum oxide, zinc oxide and silica may be vapor-deposited onto these substrate layers so that they will have gas barrier properties.

One or both sides of the substrate layer may be given surface activating treatment such as corona treatment, flame treatment, plasma treatment, undercoat treatment and primer coating treatment so that its adhesion with the laminated film of the present invention will be improved. The thickness of the substrate layer is normally 5 to 1,000 \( \mu \text{m} \), preferably 9 to 100 \( \mu \text{m} \).

Specific examples of the laminates comprising such substrate layer and the laminated film of the present invention include biaxially-oriented polyethylene terephthalate film (PET)/laminated film [laminating layer thermoplastic resin (D)/intermediate layer comprising the adhesive compound composition/heat-sealing layer comprising thermoplastic resin (E)], PET/PET/laminated film, PET/aluminum foil/laminated film, PET/deposited
PET/laminated film, transparent deposited PET/ biaxially-oriented polyamide film (ONY)/laminated film, ONY/laminated film, ONY/ethylene-vinyl alcohol copolymer film/laminated film, and transparent deposited PET/ONY/laminated film. In laminating the substrate layer and the laminated film for such laminates, various methods known to the public can be used as mentioned above, including a method in which they are laminated by dry lamination using a urethane-based adhesive for dry lamination and a method in which they are laminated by extruding high-pressure-process low density polyethylene, etc. Further, a printing layer may be included in the laminates between the substrate layer and the laminated film.

Method for manufacturing the laminated film:

The laminated film of the present invention may be manufactured by various methods known to the public.

Examples of the manufacturing methods include what is called sandwich laminating method in which a film to be used as the laminating layer made of thermoplastic resin (D) and a film to become the heat-sealing layer made of thermoplastic resin (E) are produced separately in advance, and the film to become the laminating layer and the film to be used as the heat-sealing layer are laminated by melting and extruding the adhesive compound composition to form a laminate; what is called co-extrusion laminating method in which a film to become the laminating layer made of thermoplastic resin (D) and a film to become the heat-sealing layer made of thermoplastic resin (E) are produced separately in advance, and thermoplastic resin (E) and the adhesive compound composition, or thermoplastic resin (D) and the adhesive compound composition, are co-extruded to laminate the adhesive compound composition and the aforesaid films to form a laminate; what is called dry-lamination method in which a film to become the laminating layer made of thermoplastic resin (D), a film to become the heat-sealing layer made of thermoplastic resin (E) and a film made of the adhesive compound composition are produced separately in advance, and the film to become the laminating layer made of thermoplastic (D) and the film to become the heat-sealing layer made of thermoplastic resin (E) are laminated to form a laminate; and what is called co-extrusion method in which thermoplastic resin (D), thermoplastic resin (E) and the adhesive compound composition are co-extruded by use of a three-or-more multi-layer die, so that the adhesive compound composition will form the intermediate layer.

Out of these manufacturing methods, the co-extrusion method is most preferable in that it will allow the thickness of each layer to be controlled uniformly with high accuracy.

The co-extrusion method may be the casting method using a flat die such as a T die or the blown film method using a circular die.

Container enclosure materials:

The container enclosure material of the present invention comprises the aforesaid laminated film. In using the aforesaid laminated film of the present invention for an enclosure material, the laminated film may be used singly for the
enclosure material, or laminated onto the aforesaid substrate layer so that the heat-sealing layer of the laminated film will be used as the internal surface (fusion layer).

As a specific structure of such container enclosure material, a laminate comprising the aforesaid substrate layer and the laminated film of the present invention may be used.

Containers known to the public may be used as the containers to which the container enclosure material is applicable so long as they can be heat-fused. Specific examples include those containers made of propylene-based polymer, ethylene-based polymer, styrene-based polymer, polyester, polyamide or other thermoplastic resin which have the form of sheet, tray, cup, bottle, etc. and those containers made of papers laminated on their inside with ethylene-based polymer laminates, which have the aforesaid forms. Containers made of thermoplastic resin may be either single-layer or multi-layer laminates made of two or more thermoplastic resins.

Containers made of propylene-based polymer as used in combination with the container enclosure material of the present invention is superior because they have heat resistance and a proper degree of flexibility.

Packages:

The packages of the present invention are packages comprising the aforesaid laminated film at least on one side. Examples of these packages include a four-way-sealed package which is obtained by filling contents into a three-way-sealed package formed by at least one laminated film on one side or two sheets of laminated film made of the substrate layer laminated onto the laminating surface of the laminated film as required and then sealing the mouth of the package, and a three-way-sealed package which is obtained by filling the contents into a two-way-sealed package with the heat-sealed mouth and then sealing the side after folding the laminated film.

Examples of the packages of the present invention include packages formed by filling contents into those containers having a heat-sealable sealing layer, such as films, sheets, trays, cups and bottles, and then sealing the top (mouth) of the packages with a container enclosure material (sealing material) comprising the aforesaid laminated film.

Furthermore, a laminated film in the form of tape may be attached to at least one side of such section of a package of this invention as is to be opened.

Examples:

Given below is an explanation of the present invention by using examples, but the present invention is not limited in any way by any of the examples.

Method for evaluating physical properties:

(1) Heat-sealing strength:

Before measuring the heat-sealing strength, a multi-layer film was prepared by
dry-laminating the laminating layer of a corona-treated laminated film and biaxially-oriented polyethylene terephthalate film 12 μm in thickness by use of a urethane-based adhesive (available Mitsui Takeda Chemicals, Inc.: product name: Takelac A616 (50%) + Takelac A65 (3.13%) + ethyl acetate (46.87%).)

In measuring the heat sealing strength, the heat sealing layer of the laminated film and non-oriented polypropylene sheet 300 μm in thickness were heat-sealed under the conditions of seal gauge pressure of 0.2 MPa, time of 1 second and sealing width of 5 mm at each temperature by using Heat Sealer TP-701-B available from Tester Industry Co., Ltd. Further, only the top side was heated. Next, the heat-sealed test specimen was cut to a width of 15 mm, and peeled off at a pulling rate of 500 mm/min by means of a Tensillon tester RTM-100 which is available from Orientec Co., Ltd. The maximum strength as measured in the test was taken as the heat sealing strength of the specimen. The measurement value is an average of five measurements. The peeled condition of the opened heat-sealed section at the mouth as well as heat sealing strength was observed.

(2) Resealing strength:

Following measurement was performed with the biaxially-oriented polyethylene terephthalate film dry-laminated by the laminating layer of a corona-treated laminated film of the multi-layer film, which was prepared in (1). The peeled-off heat-sealed surfaces for which heat sealing strength had been measured were put together and resealed under the conditions of set temperature of 30°C, seal gauge pressure of 1.5 MPa, time of 1 second and resealing width of 10 mm by using Heat Sealer TP-701-B available from Tester Industry Co., Ltd. Both top and bottom surfaces were heated. Next, the resealed test specimen was peeled off at a pulling rate of 500 mm/min by means of a Tensillon tester RTM-100 which is available from Orientec Co., Ltd. The maximum strength as measured in the test was taken as the resealing strength of the specimen. The measurement value is an average of the measurements of the test specimens which were successfully resealed.

(3) Heat sealing strength and resealing strength at low temperature:

Following measurement was performed with the biaxially-oriented polyethylene terephthalate film dry-laminated by the laminating layer of a corona-treated laminated film of the multi-layer film, which was prepared in (1). In measuring the resealing strength at low temperature, the heat sealing layer of the laminated film and non-oriented polypropylene sheet 300 μm in thickness were heat-sealed under the conditions of seal gauge pressure of 0.2 MPa, time of 1 second, sealing width of 5 mm and temperature of 200°C by using Heat Sealer TP-701-B available from Tester Industry Co., Ltd. Further, only the top side was heated. Next, the heat-sealed test specimen was cut to a width of 15 mm, and peeled off at a pulling rate of 500 mm/min by means of a Tensillon tester RTC-1225 which is available from Orientec Co., Ltd. and equipped with constant-temperature bath TLF-R3-C available from Orientec Co. Ltd. 5 minutes after the temperature reached the specified level. The maximum strength as
measured in the test was taken as the heat sealing strength of the specimen. The measurement value is an average of five measurements. The peeled condition of the opened heat-sealed section at the mouth as well as heat sealing strength was observed.

In measuring resealing strength at low temperature, the peeled-off heat-sealed surfaces for which heat sealing strength had been measured at the specified temperature were removed from the constant-temperature bath, put together and resealed under the conditions of set temperature of 30°C, seal gauge pressure of 1.5 MPa, time of 1 second and sealing width of 10 mm by using Heat Sealer TP-701-B available from Tester Industry Co., Ltd. Both top and bottom surfaces were heated. Next, the resealed test specimen was immediately put in the constant-temperature bath TLF-R3-C available from Orientec Co. Ltd. and peeled off at a pulling rate of 500 mm/min by means of a Tensilon tester RTC-1225 which is available from Orientec Co., Ltd and equipped with constant-temperature bath TLF-R3-C available from Orientec Co. Ltd. The maximum strength as measured in the test was taken as the resealing strength of the specimen 2 min after the temperature reached the specified level. The maximum strength as measured in the test was taken as the resealing strength of the specimen. The measurement value is an average of the measurements of the test specimens which were successfully resealed.

(4) Melting point (Tm):
The melting point was measured by DSC at a heating rate of 10 degree in Celsius (°C) per min in accordance with the testing procedure based on JIS K 7121.

Raw materials:
The polymers used in the examples and relative examples are as follows:
(1) Olefin-based block copolymer elastomer (A):
(1-1) Styrene-diene block copolymer (A1)
   (i) Hydrogenated material (SEBS) of styrene-butadiene-styrene block copolymer (SBS): Available from KRATON Polymer Japan, product name: KRATON G1657, MFR: 8 g/10 min
(1-2) Olefin-noncrystalline olefin block copolymer (A2)
   (i) Styrene-ethylene • butene-crystalline olefin block copolymer (SEBC): Available from JSR, product name: DYNALON 4600P, MFR: 5.6 g/10 min (230°C, 2.16 kg, ASTM D1238)
   (ii) Crystalline olefin-ethylene • butene-crystalline olefin block copolymer (CEBC): Available from JSR, product name: DYNALON 6200P, MFR: 2.5 g/10 min (230°C, 2.16 kg, ASTM D1238)
(2) Ethylene-vinyl acetate copolymer (B)
   (i) Ethylene-vinyl acetate copolymer (40% EVA): Vinyl acetate content: 40 wt%, density: 970 kg/m³, MFR: 2 g/10 min
   (ii) Ethylene-vinyl acetate copolymer (33% EVA): Vinyl acetate content: 33 wt%, density: 960 kg/m³, MFR: 1 g/10 min
(3) Tackifier:
(i) Terpene resin: Available from Yasuhara Chemical Co., Ltd., YS Resin PX1150N, ring-and-ball softening point: 115°C
(4) Thermoplastic resin (D) (laminating layer resin):
(i) Polypropylene homopolymer (H-PP): Available from Mitsui Chemicals, Inc., Mitsui Polypro F107DV, density: 910 kg/m³, melting point: 160°C, MFR: 7 g/10 min
(5) Thermoplastic resin (E) (heat sealing layer resin)
(i) Propylene-ethylene-1-butene random copolymer (R-PP): Available from Mitsui Chemicals, Inc., Mitsui Polypro F329D, density: 910 kg/m³, melting point: 136°C, MFR: 9 g/10 min

Example 1:

H-PP was used as the resin for the laminating layer, the adhesive compound composition obtained by mixing uniformly 40 wt% of SEBS, 40 wt% of 40% EVA, 20 wt% of terpene resin (the total parts by weight of SEBS, 40% EVA and terpene is equal to 100 parts by weight) , and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer, and R-PP was used as the resin for the heat sealing layer. Each of these were charged to 3 units of 40-mm-diameter extruder for forming non-oriented three-different layer film, extruded at 180 to 230 °C through a T-die of the feed block type at 210°C, quenched with casting roll at 30 °C and treated with corona. As a result, the laminated film was obtained.

The total thickness of the laminated film was 50 μm, and the thickness of the layers was the laminating layer/adhesive compound composition intermediate layer/heat sealing layer = 15/30/5 μm.

Results of the evaluation of the laminated film are shown in Table 1.

Example 2:

The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 40 wt% of SEBS, 40 wt% of 40% EVA, 20 wt% of aliphatic hydrocarbon resin (the total parts by weight of SEBS, 40% EVA and aliphatic hydrocarbon is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 1.

Example 3:

The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 30 wt% of SEBS, 50 wt% of 40% EVA, 20 wt% of terpene resin (the total parts by weight of SEBS, 40% EVA and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate
layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 1.

Example 4:

The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 30 wt% of SEBS, 50 wt% of 40% EVA, 20 wt% of aliphatic hydrocarbon resin (the total parts by weight of SEBS, 40% EVA and aliphatic hydrocarbon resin is 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 1.

Example 5:

The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 20 wt% of SEBS, 60 wt% of 40% EVA, 20 wt% of aliphatic hydrocarbon resin (the total parts by weight of SEBS, 40% EVA and aliphatic hydrocarbon resin is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 1.

Example 6:

The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 40 wt% of SEBS, 40 wt% of 33% EVA, 20 wt% of aliphatic hydrocarbon resin (the total parts by weight of SEBS, 33% EVA and aliphatic hydrocarbon resin is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 1.
**Table 1**

<table>
<thead>
<tr>
<th>Laminating layer</th>
<th>Example 1</th>
<th>Example 2</th>
<th>Example 3</th>
<th>Example 4</th>
<th>Example 5</th>
<th>Example 6</th>
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<tbody>
<tr>
<td></td>
<td>H-PP</td>
<td>H-PP</td>
<td>H-PP</td>
<td>H-PP</td>
<td>H-PP</td>
<td>H-PP</td>
</tr>
<tr>
<td>Intermediate layer</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Block polymer(%)</td>
<td>SEBS 40</td>
<td>SEBS 40</td>
<td>SEBS 30</td>
<td>SEBS 30</td>
<td>SEBS 20</td>
<td>SEBS 40</td>
</tr>
<tr>
<td>EVA (%)</td>
<td>40%EVA 40</td>
<td>40%EVA 40</td>
<td>40%EVA 50</td>
<td>40%EVA 50</td>
<td>40%EVA 60</td>
<td>33%EVA 40</td>
</tr>
<tr>
<td>Tackifier (%)</td>
<td>Terpene resin 20</td>
<td>*3 20</td>
<td>Terpene resin 20</td>
<td>*3 20</td>
<td>*3 20</td>
<td>*3 20</td>
</tr>
<tr>
<td>Antioxidant (parts)</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
</tr>
<tr>
<td>Heat sealing layer</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
</tr>
<tr>
<td>Heat sealing temp.</td>
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<td>160°C 180°C</td>
<td>160°C 180°C</td>
<td>160°C 180°C</td>
<td>160°C 180°C</td>
<td>160°C 180°C</td>
</tr>
<tr>
<td>Heat sealing strength</td>
<td>*1 16.9 15.2</td>
<td>17.9 20.3</td>
<td>21.7 16.4</td>
<td>19.0 20.8</td>
<td>20.4 19.1</td>
<td>18.5 20.4</td>
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<tr>
<td>Mode</td>
<td>*2 B B</td>
<td>B B</td>
<td>C B</td>
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<td>B B</td>
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<td>Milk white</td>
<td>Milk white</td>
<td>No milk white</td>
<td>Milk white</td>
<td>Milk white</td>
</tr>
<tr>
<td>Resealing strength (N/15 mm)</td>
<td>1st time 5.0 4.5 4.7 5.2</td>
<td>3.6 4.1</td>
<td>3.2 4.6</td>
<td>2.5 2.5</td>
<td>2.9 2.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2nd time 3.6 3.7</td>
<td>3.3 3.6</td>
<td>3.2 3.8</td>
<td>2.5 4.4</td>
<td>2.6 1.5</td>
<td>1.6 1.8</td>
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<tr>
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<td>3.0 4.0</td>
<td>2.1 3.9</td>
<td>1.9 0.9</td>
<td>1.3 1.9</td>
</tr>
<tr>
<td></td>
<td>4th time 2.7 4.1</td>
<td>3.3 3.8</td>
<td>2.7 3.6</td>
<td>2.6 3.8</td>
<td>2.4 0.7</td>
<td>0.9 1.6</td>
</tr>
<tr>
<td></td>
<td>5th time 2.7 3.9</td>
<td>3.0 3.5</td>
<td>2.6 3.4</td>
<td>2.8 4.1</td>
<td>1.9 0.7</td>
<td>1.1 1.2</td>
</tr>
</tbody>
</table>

*1: N/15mm
*2: A. Intermediate layer/heat sealing layer peeled off
   B. Intermediate layer failed cohesively
   C. Laminating layer/intermediate layer peeled off
*3: Aliphatic hydrocarbon resin
Example 7:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 30 wt% of SEBC, 50 wt% of 40% EVA, 20 wt% of terpene resin (the total parts by weight of SEBC, 40% EVA and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.

Example 8:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 30 wt% of CEBC, 50 wt% of 40% EVA, 20 wt% of terpene resin (the total parts by weight of CEBC, 40% EVA and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.

Comparative Example 1:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 80 wt% of 40% EVA, 20 wt% of aliphatic hydrocarbon resin (the total parts by weight of 40% EVA and aliphatic hydrocarbon resin is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.

Comparative Example 2:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 80 wt% of SEBS, 20 wt% of terpene resin (the total parts by weight of SEBS and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.

Comparative Example 3:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 80 wt% of SEBC, 20 wt% of terpene resin (the total parts by weight of SEBC and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.

Comparative Example 4:
The laminated film was obtained by the same method as described in Example 1 except that a resin obtained by mixing uniformly 80 wt% of CEBC, 20 wt% of terpene resin (the total parts by weight of CEBC and terpene is equal to 100 parts by weight) and 0.1 part by weight of phenol-based antioxidant in a twin-screw extruder was used as the intermediate layer comprising the adhesive compound composition.

Results of the evaluation of the laminated film are shown in Table 2.
<table>
<thead>
<tr>
<th>Laminating layer</th>
<th>Example 7</th>
<th>Example 8</th>
<th>Comparative Example 1</th>
<th>Comparative Example 2</th>
<th>Comparative Example 3</th>
<th>Comparative Example 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Block polymer (%)</td>
<td>SEBC 30</td>
<td>CEBC 30</td>
<td>H-PP</td>
<td>SEBS 80</td>
<td>SEBC 80</td>
<td>CEBC 80</td>
</tr>
<tr>
<td>EVA (%)</td>
<td>40% EVA 50</td>
<td>40% EVA 50</td>
<td>40% EVA 80</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tackifier (%)</td>
<td>Terpene resin 20</td>
<td>Terpene resin 20</td>
<td>*3 20</td>
<td>Terpene resin 20</td>
<td>Terpene resin 20</td>
<td>Terpene resin 20</td>
</tr>
<tr>
<td>Antioxidant (parts)</td>
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<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
<td>phenol-based 0.1</td>
</tr>
<tr>
<td>Heat sealing layer</td>
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<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
<td>R-PP</td>
</tr>
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<td>Heat sealing temp.</td>
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<td>160°C 180°C</td>
<td>180°C 180°C</td>
<td>180°C 180°C</td>
<td>180°C 180°C</td>
<td>180°C 180°C</td>
</tr>
<tr>
<td>Heat sealing strength</td>
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<td>20.5 24.9</td>
<td>13.9</td>
<td>32.2</td>
<td>25.3</td>
</tr>
<tr>
<td>Mode</td>
<td>*2</td>
<td>C B</td>
<td>B B</td>
<td>A</td>
<td>A, C</td>
<td>C</td>
</tr>
<tr>
<td>Peeled condition</td>
<td>No milk white</td>
<td>Milk white</td>
<td>Milk white</td>
<td>Milk white</td>
<td>No milk white</td>
<td>No milk white</td>
</tr>
<tr>
<td>Resealing strength (N/15 mm)</td>
<td>1st time</td>
<td>2.8 3.1</td>
<td>4.1 2.3</td>
<td>0.7</td>
<td>0.2</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td>2nd time</td>
<td>2.2 2.5</td>
<td>2.8 1.7</td>
<td>0.5</td>
<td>0.2</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>3rd time</td>
<td>1.8 1.9</td>
<td>3.3 1.9</td>
<td>0.3</td>
<td>0.1</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>4th time</td>
<td>1.7 2.0</td>
<td>2.4 2.1</td>
<td>0.4</td>
<td>0.2</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>5th time</td>
<td>1.8 1.9</td>
<td>2.7 2.5</td>
<td>0.3</td>
<td>0.1</td>
<td>0.6</td>
</tr>
</tbody>
</table>

*1: N/15mm
*2: A. Intermediate layer/heat sealing layer peeled off
   B. Intermediate layer failed cohesively
   C. Laminating layer/intermediate layer peeled off
   D. Broken seal
*3: Aliphatic hydrocarbon resin
Example 9:
Results of evaluation under low temperature (10°C, 5°C, -10°C) of the laminated film used in Example 1 are shown in Table 3.

<table>
<thead>
<tr>
<th>Laminating layer</th>
<th>Example 9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Block polymer</td>
<td>SEBS</td>
</tr>
<tr>
<td>(%) EVA</td>
<td>40%EVA</td>
</tr>
<tr>
<td>Tackifier (%)</td>
<td>Terpene resin</td>
</tr>
<tr>
<td>Antioxidant (parts)</td>
<td>phenol-based</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Heat sealing layer</th>
<th>R-PP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat sealing temp.</td>
<td>200°C</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Heat sealing strength</th>
<th>17.9</th>
<th>15.4</th>
<th>14.8</th>
<th>18.1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mode</td>
<td>B</td>
<td>B</td>
<td>B</td>
<td>B</td>
</tr>
<tr>
<td>Condition of peeled surface</td>
<td>Milk white</td>
<td>Milk white</td>
<td>Milk white</td>
<td>Milk white</td>
</tr>
<tr>
<td>Ambient temp.</td>
<td>21°C</td>
<td>10°C</td>
<td>5°C</td>
<td>-10°C</td>
</tr>
<tr>
<td>Resealing strength</td>
<td>5.4</td>
<td>5.0</td>
<td>5.8</td>
<td>6.8</td>
</tr>
<tr>
<td>(N/15 mm)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1st time</td>
<td>4.5</td>
<td>4.9</td>
<td>6.1</td>
<td>7.3</td>
</tr>
<tr>
<td>2nd time</td>
<td>4.3</td>
<td>4.6</td>
<td>6.2</td>
<td>4.8</td>
</tr>
<tr>
<td>3rd time</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*1: N/15mm
*2: A.: Intermediate layer/heat sealing layer peeled off
       B.: Intermediate layer failed cohesively
       C.: Laminating layer/Intermediate layer peeled off

As can be seen clearly from Examples as shown in Table 1 and Table 2, the laminated film formed by using the adhesive compound composition comprising olefin-based block copolymer elastomer (A), ethylene-vinyl acetate copolymer (B) having a vinyl acetate content of 25 to 80 wt% and tackifier (C) as the intermediate layer is used for the enclosures, etc. of packaging materials, such packaging materials show appropriate heat seal strength (strength at the time of opening) and adequate strength for the adhesive compound composition layer to fail cohesively when they are opened and be sealed again repeatedly. Furthermore, since it is possible by selecting heat sealing conditions to allow the interface at which the adhesive compound composition layer has once failed cohesively to become milk white, this feature is instrumental in preventing anyone from tampering from the packages.

By contrast, the packaging materials not using olefin-based block copolymer elastomer (A) as the intermediate layer is low in resealing strength, and cannot evidently be resealed again repeatedly (Comparative Example 1). On the other hand, a laminated film which does not use ethylene-vinyl acetate copolymer (B) or use SEBS or CEBC as olefin-based block copolymer elastomer (A) shows high heat seal strength so that its easy-to-open properties become poor, its reseal strength becomes low (Comparative Example 2) and its seal is broken at the time of opening.
(Comparative Example 4). Further, a laminated film made of a composition using SEBC as olefin-based block copolymer elastomer (A) cannot be resealed repeatedly (Comparative Example 3). Therefore, it is evident that none of them is suitable for a packaging material that has easy-to-open properties and resealability.

Effect of the invention:

The adhesive compound composition of the present invention has an excellent characteristic in that it has both easy-to-open properties and an appropriate degree of tackiness suitable for a resealable packaging material. If a film formed by laminating a laminating layer made of a thermoplastic resin and a heat-sealing layer made of a thermoplastic resin with said composition used as the intermediate layer is used as a packaging material by making use of said characteristic, such packaging material will show excellent heat sealability and provide a package which will be easy to open when it is sealed by heat sealing (heat fusion). In addition to that, when such package is opened, the heat sealing layer will break, while the heat sealing layer and the adhesive compound composition layer will be delaminated, or the adhesive compound composition layer will fail cohesively or will also break with the result that the adhesive compound composition layer and the laminating layer will be delaminated, causing the intermediate layer or the adhesive compound composition layer to be exposed to the opened surface. The characteristic of such adhesive compound composition layer is such that it will show adequate adhesion even when it is pressed by the tip of a finger because of its excellent self-adhesive properties and its self-adhesive properties will not decline even if it is peeled off and bonded repeatedly at low temperature (after being put into and taken out of a refrigerator) as well as at normal temperature (room temperature).

Furthermore, the laminated film of the present invention allows a package using it to be determined visually to be a one which is once opened and then resealed or a one which has not been opened by properly selecting the type or composition ratio of the polymer, etc. used in the adhesive compound composition, the thermoplastic plastic resin used for the laminating layer and the thermoplastic resin used for the heat sealing layer so that when the heat-sealed section of the laminated layer used in the package is peeled off, the heat seating layer will be broken, the adhesive compound composition of the intermediate layer will fail cohesively at the same time, and the peeled-off section will turn milk-white and the resealed section will remain in such milk-white condition even after it is resealed. This allows anyone to judge from the appearance of a package using the laminated film if it has been resealed or not yet been opened.

Possibility of Industrial Use:

The adhesive compound composition has excellent self-adhesive properties and declines in self-adhesive properties even after repeated peeling-off and bonding. For this reason, the adhesive compound composition provides a packaging material showing excellent sealability when it is used in the intermediate layer of film made of thermoplastic resin.

The laminated film of the present invention is such that after two heat sealing layers
(heat-fusion layers) made of the aforesaid thermoplastic resin (E), or such heat sealing layer and a film or container having a heat sealing layer that is heat-sealable, are heat-sealed (heat-fused), the heat sealing layer is broken with the result that the adhesive compound composition intermediate layer having self-adhesive properties will be exposed to the peeled-off surface, or preferably the adhesive compound composition layer will fail cohesively, thus being exposed to the peeled-off surfaces of both layers, when the aforesaid heat-sealed section is peeled off (opened). Because of this, the laminated film has the characteristic of being resealed when such peeled-off surface is pressed by a finger. In addition, the laminated film has the advantage of showing no decline in resealing strength after it is opened and bonded repeatedly.

Because of the aforesaid characteristic, the laminated film of the present invention is well suited for use as a packaging material, particularly an enclosure material for those sheets, trays, cups and bottles made of propylene-based polymer and can be used suitably for packages of snacks such as rice crackers and potato chips and food such as jelly, milk, yogurt, pudding, soybean curd and lactic acid beverage, blister packages and other materials for packaging pharmaceuticals, medical equipment, articles of daily use, general merchandise.
CLAIMS

1. An adhesive compound composition comprising an olefin-based block copolymer elastomer (A), an ethylene-vinyl acetate copolymer (B) having a vinyl acetate content of 25 to 80 wt% and a tackifier (C).

2. An adhesive compound composition as claimed in claim 1, wherein said olefin-based block copolymer elastomer (A) is 10 to 80 wt%, said ethylene-vinyl acetate copolymer having a vinyl acetate content of 25 to 80 wt% (B) is 10 to 80 wt% and said tackifier (C) is 10 to 40 wt%, with (A) + (B) + (C) being 100 wt%.

3. An adhesive compound composition as claimed in claim 1 or claim 2, wherein said olefin-based block copolymer elastomer (A) is a styrene-diene block copolymer (A1) and/or an olefin-noncrystalline olefin block copolymer (A2) having at least one noncrystalline olefin polymer block and a crystalline olefin polymer block.

4. An adhesive compound composition as claimed in claim 3, wherein said styrene-diene block copolymer (A1) is a hydrogenated material.

5. An adhesive compound composition as claimed in claim 4, wherein said hydrogenated material of said styrene-diene block copolymer (A1) is a hydrogenated material of any one or more block copolymers selected from among a styrene-butadiene block copolymer, a styrene-butadiene-styrene block copolymer and a styrene-isoprene-styrene block copolymer.

6. An adhesive compound composition as claimed in claim 3, wherein said olefin-noncrystalline olefin block copolymer (A2) is either a styrene-ethylene butene-crystalline olefin block copolymer or a crystalline olefin-ethylene butene-crystalline olefin block copolymer.

7. An adhesive compound composition as claimed in any of claims 1 through 6, wherein said tackifier (C) is any one or more of a rosin derivative, a terpene-based resin, petroleum resin and hydrogenated petroleum resin.

8. A laminated film comprising a heat-sealing layer made of a thermoplastic resin (E) which is laminated onto one side of a laminating layer made of a thermoplastic resin (D) via an intermediate layer made of an adhesive compound composition as claimed in any of claims 1 through 7.

9. A laminated film as claimed in claim 8, wherein said thermoplastic resin (D) is a propylene-based polymer.

10. A laminated film as claimed in claim 8, wherein said thermoplastic resin (E) is a propylene-α-olefin random copolymer.
11. A laminated film as claimed in any one of claims 8 through 10, wherein after the heat-sealing layer made of thermoplastic resin (E) of said laminated film is heat-sealed with another heat-sealable thermoplastic resin, the heat-sealing layer fails and at the same time said intermediate layer comprising the adhesive compound composition fails cohesively when such heat-sealed section is peeled, with the result that the adhesive compound composition is exposed on both sides of such peeled heat-sealed section.

12. A container enclosure material comprising a laminated film as claimed in any one of claims 8 through 11.

13. A package formed by heat-sealing the heat-sealing layer of a laminated film as claimed in any one of claims 8 through 11.

14. A package formed by heat-sealing the heat-sealing layer of a laminated film as claimed in any one of claims 8 through 11 and another heat-sealable thermoplastic resin layer.

15. A package formed by heat-sealing the top surface of a container with a container enclosure material as claimed in claim 12.

16. A package as claimed in any one of claims 13 through 15, wherein when said package is opened, the heat-sealing layer made of thermoplastic resin (E) of the laminated film fails and at the same time the intermediate layer comprising an adhesive compound composition fails cohesively when such heat-sealed section is peeled, with the result that the adhesive compound composition is exposed on both sides of the failed heat-sealed section.