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(54) Title: MULTILAYERED POLYMERIC FILM FOR HOT MELT ADHESIVE LABELING AND LABEL STOCK AND LABEL THEREOF

(B)
(A)
(C)

(57) Abstract: A polymeric film for labeling articles comprises a core layer (A), a print skin layer (B) comprising a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers, and an adhesive skin layer (C) comprising a polypropylene homopolymer, where the polymeric film is oriented. An adhesive containing label stock and an adhesive label die-cut from the adhesive containing label stock comprise the polymeric film which is especially useful for pressure sensitive hot melt adhesive applications.



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TITLE: MULTILAYERED POLYMERIC FILM FOR HOT MELT ADHESIVE LABELING AND LABEL STOCK AND LABEL THEREOF**Cross-References to Related Applications**

This application claims the benefit of U. S. Provisional Application No. 60/805,231 filed June 20, 2006.

Field of the Invention

This invention is related to a multilayered polymeric film for labeling articles and is further related to an adhesive containing label stock and a label comprising the polymeric film. The film, label stock and label are especially useful with a hot melt adhesive.

Background of the Invention

Hot melt adhesives provide a convenient and economical way to label articles of commerce such as glass, metal and plastic containers for consumer and industrial products. Hot melt adhesives are easy to handle in their solid form, generally have an indefinite shelf life, are environmentally friendly and cost effective since they do not involve solvents, and are able to quickly form adhesive bonds without any supplementary processing. One disadvantage of hot melt adhesives is their tendency to damage a label. Various relatively low molecular weight additives generally present in hot melt adhesives can migrate into a polymeric film of a label and result in staining, swelling and distortion of the label which detract from its function and appearance. This additive migration can also gradually result in the hot melt adhesive losing tack or adhesiveness especially at high service temperatures which can result in partial or complete detachment of the label. Hot melt adhesives can also cause thermal distortion of labels which lack heat resistance.

The polymeric film of the present invention minimizes or eliminates migration of hot melt adhesive additives and is heat resistant so that derivative labels are not damaged and remain attached to a container even at

high service temperatures. The inventive polymeric film further provides the performance that is required to produce high quality labels including printability, die-cuttability, sufficient stiffness for dispensability, and conformability to an article or container.

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Summary of the Invention

The present invention involves a polymeric film for labeling articles that comprises (A) a core layer having an upper surface and a lower surface and comprising a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers; (B) a print skin layer comprising a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers wherein the print skin layer overlies the upper surface of the core layer; and (C) an adhesive skin layer comprising a polypropylene homopolymer wherein the adhesive skin layer has an upper surface and a lower surface wherein the upper surface of the adhesive skin layer underlies the lower surface of the core layer; wherein the composition of layer (C) is different from the composition of layer (A), and the polymeric film is oriented.

In an embodiment of the invention, the core layer (A) of the polymeric film further comprises a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers.

In another embodiment of the invention, an adhesive containing label stock for use in adhesive labels comprises the above described polymeric film of this invention comprising layers (A), (B) and (C); (D) an adhesive layer having an upper and a lower surface wherein the upper surface of the adhesive layer is adhesively joined to the lower surface of layer (C); and (E) a release liner wherein the release liner is releasably adhered to the lower surface of the adhesive layer (D).

In a further embodiment of this invention an adhesive label is die-cut from the above described adhesive containing label stock comprising layers (A), (B), (C), (D) and (E).

5 **Brief Description of the Drawings**

Figure 1 is a cross sectional view of a polymeric film of the present invention.

Figure 2 is a cross sectional view of an adhesive containing label stock of the present invention.

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Detailed Description of the Invention

The polymeric film of this invention comprises (A) a core layer having an upper surface and a lower surface, (B) a print skin layer that overlies the upper surface of the core layer (A), and (C) an adhesive skin layer that underlies the lower surface of the core layer (A). The terms overlie and underlie mean that the print skin layer and adhesive skin layer can be bonded to or in direct contact with the core layer or that one or more intermediate layers such as, for example, a tie layer can be between a skin layer and the core layer.

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(A) Core Layer

The core layer (A) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene having a density of 0.94 g/cm³ or higher, or a mixture of any of the foregoing polymers. A mixture as used throughout this application comprises two or more components of the same kind, or one or more components of the same kind and one or more components of one or more different kinds. For example the mixture of polymers in layer (A) can comprise two or more polypropylene homopolymers, two or more polypropylene copolymers, two or more high density polyethylenes, or one or more polypropylene homopolymers and/or one or more polypropylene copolymers and/or one or more high density polyethylenes. The polypropylene homopolymers and polypropylene copolymers and high density polyethylenes provide a rigid film for applications requiring a rigid film. Polypropylene homopolymers comprise thermoplastic

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polymers having a density of about 0.9 g/cm^3 and a melt index (also known as a melt flow rate) of about 0.5 to 40 g/10 minutes as measured by ASTM Standard D1238. Polypropylene homopolymers can be prepared from by polymerizing propylene monomer using either a Ziegler-Natta or metallocene catalyst, and are available from various resin suppliers including ExxonMobil, Huntsman and Dow Chemical. The polypropylene homopolymer can comprise a crystalline polypropylene homopolymer comprising an isotactic polypropylene homopolymer, a syndiotactic polypropylene homopolymer, or a mixture of any of the foregoing polypropylene homopolymers. The mixture of polypropylene homopolymers can be two or more isotactic polypropylene homopolymers, two or more syndiotactic polypropylene homopolymers, or one or more isotactic polypropylene homopolymers and one or more syndiotactic polypropylene homopolymers. Both isotactic and syndiotactic polypropylene homopolymers are highly crystalline having little or no solubility in toluene and having respectively melting points of about $150\text{-}175^\circ\text{C}$ and about $120\text{-}135^\circ\text{C}$. An isotactic polypropylene homopolymer can have an isotactic stereoregularity based on ^{13}C nuclear magnetic resonance (NMR) spectroscopy as described in US Patent No. 5476914 of greater than about 70%, 80% or 90%. A syndiotactic polypropylene homopolymer can have a syndiotactic stereoregularity based on ^{13}C NMR spectroscopy as described in US Patent No. 5476914 of greater than about 70%, 80% or 90%. An isotactic polypropylene homopolymer can be prepared from Ziegler-Natta or metallocene catalysts and is available from resin suppliers including ExxonMobil, Huntsman, BASF, Amoco, Chisso and Solvay. A useful polypropylene homopolymer is Huntsman P4G4K-173X, which is a nucleated polypropylene homopolymer having a density of 0.9 g/cm^3 and an ASTM D1238 melt index at $230^\circ\text{C}/2.16 \text{ kg}$ of 12 g/10 minutes. A syndiotactic polypropylene homopolymer, which is less crystalline than an isotactic polypropylene homopolymer, can be prepared with stereorigid metallocene catalysts as described in US Patent No. 5476914 and is available from resin supplier Atofina from the Finaplas[®] series of syndiotactic polypropylene homopolymers.

Polypropylene copolymers comprise thermoplastic polymers having a density of about or less than 0.9 g/cm^3 , a melt index of about 0.5 to 40 g/10

minutes, and a high degree of crystallinity with a melting point of about 120 to 150°C. Polypropylene copolymers can be prepared by polymerizing propylene monomer and one or more alpha-olefin comonomers using catalysts such as Ziegler-Natta and metallocene catalysts. The alpha-olefin comonomers can
5 comprise ethylene and alpha-olefins having 4 to 30 or 4 to 12 carbon atoms. The alpha-olefin comonomer content in polypropylene copolymers can range on a weight basis from 0.1 to 40%, 0.2 to 20%, 0.3 to 10%, or 1 to 7%. In an embodiment of the invention the polypropylene copolymer is a polypropylene random copolymer prepared from a Ziegler-Natta catalyst. In another
10 embodiment of the invention the polypropylene random copolymer is a propylene-ethylene copolymer containing about 1 to 7% by weight of the ethylene comonomer. Polypropylene copolymers are available from various resin suppliers including ExxonMobil, Dow Chemical and Huntsman. A useful polypropylene copolymer is Huntsman P5M4K-070X, which is a nucleated
15 polypropylene copolymer containing 3.2% by weight ethylene and having a density of 0.9 g/cm³ and an ASTM D1238 melt index at 230°C/2.16 kg of 10 g/10 minutes.

High density polyethylenes are available from various resin suppliers including Dow Chemical and Atofina. Useful high density polyethylenes are
20 Dow Chemical HDPE 12450N and HDPE 17450N which have respectively an ASTM D1238 melt index at 190°C/2.16 Kg of 12 and 17 g/10 minutes.

In an embodiment of the invention, the core layer (A) further comprises one or more additional thermoplastic polymers. The one or more thermoplastic polymers comprise other polyolefins, alkene-unsaturated
25 carboxylic acid or acid derivative copolymers, styrene-based polymers or copolymers, polyurethanes, poly(vinyl chloride)s, polycarbonates, polyamides, fluoroplastics, poly(meth)acrylates, polyacrylonitriles, polyesters, or a mixture of any of the foregoing polymers.

In an embodiment of the invention, the core layer (A) further comprises
30 a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers. The mixture can comprise two or more polyolefin elastomers, two or more alkene-unsaturated carboxylic acid or acid derivative copolymers, or one or more polyolefin elastomers and one or more alkene-unsaturated carboxylic acid or

acid derivative copolymers. The polyolefin elastomers and alkene-unsaturated carboxylic acid or acid derivative copolymers in combination with the polypropylene and high density polyethylene polymers provide a more conformable film for applications requiring conformability.

5 The polyolefin elastomer comprises an elastomeric homopolymer or copolymer of C₂-C₃₀ olefin monomers or a mixture of any of the foregoing polyolefin elastomers. Polyolefin elastomers contain both hard or crystalline and soft or amorphous segments. Polyolefin elastomers are intermediate between thermoplastic polymers and vulcanized rubbers based on crystallinity
10 and flexibility. Polyolefin elastomers are more crystalline and less flexible than vulcanized rubbers, and are typically less crystalline and lower melting and more flexible than thermoplastic polymers such as polypropylene homopolymers or copolymers which are highly crystalline. Polyolefin elastomers can have a density of about 0.84 to 0.935 g/cm³, a melt index of
15 about 0.5 to 40g/10 minutes, a melting point of about 35 to 160°C, about 35 to 120°C, or about 40 to 110°C, and an ASTM D638 elongation at yield of greater than 40% or 60% or 130% or 200%. Polyolefin elastomers can generally be prepared by polymerizing an olefin monomer or a mixture of two or more different olefin monomers to form respectively a homopolymeric
20 polyolefin elastomer or a copolymeric polyolefin elastomer also known as an olefin copolymer elastomer. Olefin monomers can comprise an alkene having a single double bond such as for example ethylene, an aliphatic polyene having two or more double bonds to include both conjugated and nonconjugated alkadienes such as, for example, 1,3-butadiene and
25 1,4-pentadiene, or mixtures of any of the foregoing olefin monomers. An alkene can comprise an internal alkene such as, for example, 2-butene, an alpha-olefin also known as a 1-alkene such as for example ethylene and propylene and 1-octene, or a mixture of any of the foregoing alkenes. Homopolymeric polyolefin elastomers can be prepared by polymerizing an
30 olefin monomer, such as, for example, propylene, into blocks of isotactic polyolefin and blocks of atactic polyolefin to form a polyolefin elastomer chain. In an embodiment of the invention a polyolefin elastomer can comprise a low density polyethylene homopolymer prepared by free radical catalysis and having a density of 0.915 to 0.925 g/cm³. Copolymeric polyolefin elastomers

can be prepared by polymerizing an olefin monomer and one or more olefin comonomers using a metallocene catalyst where the olefin comonomer content of the elastomer can be on a mole % basis about 1.5-30%, 2-25%, 2.5-20%, or 5-20%. In embodiments of the invention, the olefin copolymer elastomer comprises two or more alpha-olefin monomers, or is prepared from two or more alpha-olefin monomers having 2-30, 2-20, 2-12, or 2-8 carbon atoms. In other embodiments of the invention the copolymeric polyolefin elastomer comprises an ethylene-alpha-olefin copolymer elastomer such as, for example, an ethylene-alpha-olefin copolymer elastomer where the alpha-olefin can comprise 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-octene, or a mixture of any of the foregoing alpha-olefins; a propylene-alpha-olefin copolymer elastomer; or a mixture of any of the foregoing copolymer elastomers. In an embodiment of the invention, the polyolefin elastomer comprises a medium density polyethylene, a linear low density polyethylene, a very low density polyethylene, a low density polyethylene prepared by free radical catalysis, a polyethylene plastomer, a polypropylene copolymer elastomer, or a mixture of any one of the foregoing polymers. Polyolefin elastomers are available from several resin suppliers and include the Exact[®] series and the Vistamaxx[®] series of elastomers from ExxonMobil and the Affinity[®] series and Versify[®] series of elastomers from Dow. Useful polyolefin elastomers include the ExxonMobil Exact[®] series of polyethylene plastomers which are prepared by polymerizing ethylene and a minor amount of 25 mole % or less of an alpha-olefin comonomer using a metallocene catalyst such as Exact[®] 8203 which has an octene comonomer and a density of 0.882 g/cm³ and an ASTM D1238 melt index at 190°C/2.16 kg of 3 g/10 minutes. Useful polyolefin elastomers also include the Dow Chemical Versify[®] series of polypropylene copolymer elastomers which are prepared by polymerizing propylene with a minor amount of ethylene such as Versify[®] 3200 which has a density of 0.876 g/cm³ and an ASTM D1238 melt index at 190°C/2.16 kg of 8 g/10 minutes.

In an embodiment of the invention, the alkene-unsaturated carboxylic acid or acid derivative copolymer comprises an alkene-unsaturated carboxylic acid copolymer, an alkene-unsaturated carboxylate ester copolymer, a metal salt of an alkene-unsaturated carboxylic acid copolymer, or a mixture of any of

the foregoing copolymers. The alkene-unsaturated carboxylic acid or acid derivative copolymer can comprise a copolymer comprising an alkene monomer and an unsaturated carboxylic acid or acid derivative comonomer where the alkene-unsaturated carboxylic acid or acid derivative copolymer can be prepared from the alkene monomer and the unsaturated carboxylic acid or acid derivative comonomer. The alkene monomer can comprise an internal alkene such as 2-butene and/or an alpha-olefin such as ethylene, and can have 2 to 30 carbon atoms. The unsaturated carboxylic acid or acid derivative comonomer can comprise unsaturated carboxylic acids such as, for example, methacrylic acid and acrylic acid, unsaturated carboxylic acid esters such as, for example, vinyl acetate and ethyl acrylate and ethyl methacrylate, or a mixture of any of the foregoing comonomers. The comonomer can be present in the copolymer on a weight basis at 1-40%, 4-30%, or 6-25%. In several embodiments of the invention, the alkene-unsaturated carboxylic acid or acid derivative copolymer can comprise an ethylene-vinyl acetate copolymer, an ethylene-alkyl (meth)acrylate copolymer, an ethylene-(meth)acrylic acid copolymer, a metal salt of an ethylene-(meth)acrylic acid copolymer (also known as an ionomer), and a mixture of any of the foregoing copolymers. Alkene-unsaturated carboxylic acid or acid derivative copolymers can have a density of 0.91-0.96 g/cm³, a melt index of 0.5 to 40 g/10 minutes, a melting point of 35-160°C or 35-120°C or 40-110°C, and are available from resin suppliers including AT Plastics and DuPont. Useful alkene-unsaturated carboxylic acid or acid derivative copolymers include A. T. Plastics Ateva[®] series of ethylene-vinyl acetate copolymers such as Ateva[®] 1231 which contains 12 wt. % vinyl acetate and has a density of 0.932 g/cm³ and an ASTM D1238 melt index at 190°C/2.16 kg of 3 g/10 minutes and Ateva[®] 1821 which contains 18 wt. % vinyl acetate and has a density of 0.938 g/cm³ and an ASTM D1238 melt index at 190°C/2.16 kg of 3 g/10 minutes. The DuPont Surlyn[®] series of ionomer resins which are metal salts of ethylene-methacrylic acid copolymers such as Surlyn[®] 1601 which is a sodium salt are also useful.

In an embodiment of the invention, the amount of the polyolefin elastomers and alkene-unsaturated carboxylic acid or acid derivative copolymers present in the core layer (A) based on the weight of the core layer (A) can be at least 10%, 10 to 50%, 10 to 40%, or 10 to 30% by weight. In an

embodiment of the invention, the amount of the polypropylene homopolymers and polypropylene copolymers and high density polyethylenes present in the core layer (A) based on the weight of the core layer (A) can be up to 90%, 50 to 90%, 60 to 90%, or 70 to 90% by weight. The presence of a polyolefin elastomer and/or alkene-unsaturated carboxylic acid or acid derivative copolymer in the core and/or print skin layers of the polymeric film can improve performance of the polymeric film in terms of increasing tear resistance, impact resistance, conformability to a container, and printability of a derivative label.

10 In an embodiment of the invention, the core layer (A) further comprises one or more additives as described in US Patent No. 6821592. The one or more additives comprise a nucleating agent, an antiblock agent, a processing aid, a slip agent, an antistatic agent, a pigment, a cavitating agent, an inorganic filler, an antioxidant, or a mixture of any of the foregoing additives.

15 The additives can be introduced into the film or a layer of the film as part of commercially available resins supplied by resin manufacturers where the resin comprises at least one thermoplastic polymer and one or more additives. Alternatively, the additives can be introduced into the film or a layer of the film as part of commercially available additive concentrates supplied by additive

20 concentrate manufacturers where the additive concentrate comprises one or more additives in a thermoplastic polymer matrix or carrier. The nucleating agent can comprise an inorganic or mineral nucleating agent such as, for example, a silica, an organic nucleating agent such as a metal salt of a carboxylic acid or a beta-nucleating agent, or a mixture of any of the foregoing

25 nucleating agents. The nucleating agent can be present in one or more layers of the polymeric film of this invention at a range of 100 to 10,000 or 200 to 5,000 or 300 to 1,000 ppm (parts per million) by weight based on the weight of the layer. The nucleating agent is generally introduced into a layer of the film as part of a thermoplastic polymer as obtained from a resin supplier. In

30 several embodiments of the invention, a nucleating agent is present in one layer of the film which can comprise the core layer or one of the skin layers, or is present in any combination of two or more layers of the film. The antiblock agent can comprise a concentrate of about 3 to 80% by weight of an inorganic mineral in a thermoplastic polymer matrix such as for example 5% by weight

of an amorphous silica in a polyolefin matrix. The antiblock agent can be present in one or more layers of the polymeric film of this invention at a range of about 100 to 10,000 or 200 to 5,000 or 300 to 1,000 ppm by weight based on the weight of the layer. Antiblock agents as well as slip agents, processing aids and antistatic agents provide improvement in performance to a film and derivative label due to surface properties. Consequently when present in the film, these additives are generally present in one or both of the skin layers. The pigment can comprise an inorganic pigment comprising titanium dioxide, calcium carbonate, talc, an iron oxide, a carbon black, or a mixture of any of the foregoing inorganic pigments; an organic pigment; or a mixture of any of the foregoing pigments. The pigment can comprise a concentrate of about 20 to 80% by weight of an inorganic pigment and/or organic pigment in a thermoplastic matrix. The pigment concentrate can be present in one or more layers of the polymeric film of this invention at a range of about 0.5 to 20% by weight based on the weight of the layer to provide color to and/or opacify the film. An opaque film will generally have an opacity of at least 70%, at least 75%, or at least 80%. The pigment concentrate is generally in the core layer when present in the film.

(B) Print Skin Layer

The print skin layer (B) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers. The polymers of the print skin layer are described hereinabove for the core layer (A). The polyolefin elastomers and alkene-unsaturated carboxylic acid or acid derivative copolymers when present in the print skin layer improve performance of the polymeric film to include the printability of a derivative label. The composition of the print skin layer (B) is different from the composition of the core layer (A) because the print skin layer and the core layer provide different functions and performance for the film.

In several embodiments of the invention, the print skin layer (B) comprises, based on the weight of the print skin layer, at least 10%, 10 to

50%, 10 to 40%, or 10 to 30% by weight of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers. In several embodiments of the invention the print skin layer (B) comprises, based on the weight of the print skin layer, up to
5 90%, 50 to 90%, 60 to 90%, or 70 to 90% by weight of a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers.

In an embodiment of the invention, the print skin layer (B) further comprises one or more additional thermoplastic polymers as described
10 hereinabove for the core layer (A).

In an embodiment of the invention, the print skin layer (B) further comprises one or more additives or additive concentrates as described hereinabove for the core layer (A). In another embodiment of the invention the print skin layer comprises a nucleating agent, an antiblock agent, a
15 processing aid, a slip agent, an antistatic agent, an antioxidant or a mixture of any of the foregoing additives.

(C) Adhesive Skin Layer

The adhesive skin layer (C) comprises a polypropylene homopolymer
20 wherein the composition of the adhesive skin layer (C) is different from the composition of the core layer (A). Because the adhesive skin layer and the core layer provide different functions and performance for the film, the composition of the adhesive skin layer is different from the composition of the core layer. The composition of layer (C) can be different from the composition
25 of layer (A) based on (i) the type and/or amount of polypropylene polymer, (ii) the type and/or amount of one or more additional thermoplastic polymers and/or polyolefin elastomers and/or alkene-unsaturated carboxylic acid or acid derivative copolymers as described hereinabove for the core layer (A), (iii) the type and/or amount of one or more additives as described hereinabove for
30 core layer (A), or (iv) a combination of any of the foregoing conditions (i)-(iii). A function of the adhesive skin layer is to provide a surface for an adhesive layer to be adhesively joined to the surface of the adhesive skin layer wherein the adhesive layer comprises an adhesive as described hereinbelow for the adhesive label stock. In an embodiment of the invention the adhesive skin

layer provides a surface for an adhesive layer to be adhesively joined to the surface of the adhesive skin layer wherein the adhesive layer comprises a hot melt adhesive and the adhesive skin layer eliminates or minimizes thermal distortion and migration of additives or components of the hot melt adhesive so that the inventive film and a derivative label from the film are not damaged or reduced in performance.

The polypropylene homopolymer of layer (C) comprises the polypropylene homopolymer as described hereinabove for the core layer (A). In an embodiment of the invention, the polypropylene homopolymer of layer (C) comprises an isotactic polypropylene homopolymer, a syndiotactic polypropylene homopolymer, or a mixture of any of the foregoing polypropylene homopolymers. In another embodiment of the invention the isotactic polypropylene homopolymer has an isotactic stereoregularity of greater than 70%, 80%, 90%, or 95%. In another embodiment of the invention the syndiotactic polypropylene homopolymer has an syndiotactic stereoregularity of greater than 70%, 80%, 90%, or 95%. In an embodiment of the invention the adhesive skin layer (C) comprises on a weight basis at least 70%, 80%, 90%, or 95% of a polypropylene homopolymer. In embodiments of the invention, layer (C) consists essentially of a polypropylene copolymer and at least 70% by weight of a polypropylene homopolymer or of a polypropylene copolymer, at least 70% by weight of a polypropylene homopolymer, and at least one additive or additive concentrate where the additives and additive concentrates are described hereinabove for the core layer (A) and hereinbelow for adhesive skin layer (C). In other embodiments of the invention, layer (C) consists essentially of a polypropylene copolymer and at least 80% by weight of a polypropylene homopolymer or of a polypropylene copolymer, at least 80% by weight of a polypropylene homopolymer, and at least one additive or additive concentrate. In embodiments of the invention layer (C) consists essentially of at least one polypropylene homopolymer or of at least one polypropylene homopolymer and at least one additive or additive concentrate. In several embodiments of the invention, the polypropylene copolymer has on a weight basis a comonomer content of less than 10%, less than 6%, less than 4%, or less than 2% wherein the comonomer is an olefin other than propylene.

In an embodiment of the invention, the adhesive skin layer (C) further comprises one or more additional thermoplastic polymers as described hereinabove for the core layer (A).

In an embodiment of the invention, the adhesive skin layer (C) further
5 comprises one or more additives or additive concentrates as described above for the core layer (A). In an embodiment of the invention the adhesive skin layer (C) comprises an antiblock agent, a slip agent, a processing aid, an antistatic agent, a nucleating agent, an antioxidant, or a mixture of any of the foregoing additives. In another embodiment of the invention the adhesive
10 layer (C) comprises at least one additive or additive concentrate wherein the additive or additive concentrate does not comprise a cavitating agent.

Film Construction and Processing

The layers of the multilayered polymeric film of the present invention
15 can be prepared by steps comprising extrusion of a layer from a linear or annular die, coextrusion of two or more layers from a linear or annular die, lamination of two or more previously formed layers, coating of one or more layers onto a previously formed monolayer or multilayer construction, or any combination of the foregoing steps. In an embodiment of the invention, the
20 film is prepared by coextrusion from a linear die.

The polymeric film can subsequently be oriented by stretching it biaxially in the machine direction (direction the film is advanced during film manufacture) and in the transverse direction (direction normal or perpendicular to the machine direction) or by stretching it uniaxially in the
25 machine direction or in the transverse direction. Generally the film is oriented by hot stretching it several times its length and/or width and then annealing it where hot stretching and annealing are done at or near the softening temperature of the film or from 90 to 175°C. In several embodiments of the invention, the film is oriented by stretching the film from 2 to 12 times, 2 to 10
30 times, 2 to 9 times, 3 to 9 times, or 4 to 6 times in the machine direction or in both the machine and transverse directions. In embodiments of the invention the film is oriented by stretching the film from 2 to 12 times, 2 to 10 times, 2 to 9 times, 3 to 9 times, or 4 to 6 times in the machine direction only. The oriented film can then be heat set or annealed to provide dimensional stability

to the film to prevent or minimize shrinking or distortion when exposed to elevated service application temperatures during processing to form a derivative label and in subsequent label service. Coextrusion, orientation and annealing procedures which can be used to manufacture a film are described
5 in US Patent Application Publication No. US2004/0033349.

Orienting a film in the machine direction only generally increases the tensile modulus or stiffness in the machine direction. This increased machine direction stiffness provides a label, prepared from the film, with sufficient stiffness for proper dispensability and good print registration in the labeling of
10 articles. This film, oriented in the machine direction only, generally has a substantially lower tensile modulus or stiffness in the transverse direction (TD) compared to the machine direction (MD) which provides a derivative label with sufficient flexibility for good conformability to the surface of an article to be labeled. In embodiments of the invention the TD to MD tensile modulus ratio
15 is less than 0.75, 0.65, or 0.6.

The film following manufacture, which can comprise for example coextrusion or coextrusion and orientation or coextrusion and orientation and annealing, can have a total thickness ranging from 0.5-10 mils (12.7-254 micrometers), 1-7 mils (25.4-177.8 micrometers), or 1.5-4 mils (38.1-101.6
20 micrometers). The core layer (A) of the inventive film is generally a thicker layer compared to the skin layers (B) and (C) or to any tie layers that might be present in the film. The thickness of the core layer (A) can be greater than 50%, 65 to 98%, 70 to 98% or 85 to 96% of the thickness of the film. The thickness of the skin layer (B) can be the same as or different from the
25 thickness of the skin layer (C). The thickness of the skin layers (B) and (C) can independently be less than 50%, less than 40%, 1 to 30%, 1 to 15%, 1 to 10%, or 2 to 5% of the thickness of the film. A tie layer can be present in the film between the core layer (A) and one or both of skin layers (B) and (C) to improve adhesion or bonding of the skin layer or skin layers to the core layer.
30 The tie layer can comprise any polymers, including thermoplastic polymers or polyolefin elastomers or mixtures of any of the foregoing polymers, as described hereinabove for the core layer (A) and throughout this application that improves adhesion, and can comprise a blend of a polyolefin and an alkene-unsaturated carboxylic acid or acid derivative copolymer and/or

polyolefin elastomer such as for example a blend of a polypropylene homopolymer or copolymer and an ethylene-vinyl acetate copolymer. The thickness of tie layers is generally the same as the thickness of the skin layers. The composition of any tie layer lying between a skin layer and the
5 core layer is different from the composition of the core layer and said skin layer since each of these layers provides different functions and performance for the film.

In embodiments of the invention each of the thermoplastic polypropylene homopolymers and/or copolymers present in the layers of the
10 film can independently have a melt flow rate in g/10 minutes as measured by ASTM Standard D1238 of at least 4, 4 to 35, 6 to 30, at least 8, or 8 to 25 at 230°C/2.16 Kg. Labels derived from a film comprising high melt flow rate (at least 4 or at least 8 g/10 minutes) polypropylene homopolymers and/or copolymers generally have improved die-cutability.

15 The film of the present invention comprises a print skin layer (B) which comprises at least 10% by weight of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or mixture of any of the foregoing polymers in the print skin layer based on the weight of the print skin layer. The polyolefin elastomers and alkene-unsaturated carboxylic acid
20 or acid derivative copolymers can improve printability and/or allow for improving printability in terms of ink adhesion to a label derived from the film. The film can also be treated on the surface of the print skin layer (B) or on the surfaces of both skin layers to further improve adhesion of an ink or an adhesive layer to a skin layer. Surface treatments comprise corona discharge
25 treatment, flame treatment, plasma treatment, coating with an ink- or adhesive-receptive material such as an acrylic coating, or a combination of any of the foregoing treatments.

In several embodiments of the invention a core layer (A) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density
30 polyethylene, or a mixture of any of the foregoing polymers, and the print skin layer (B) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative

copolymer, or a mixture of any of the foregoing polymers . In several more embodiments of this invention a core layer (A) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene or a mixture of any of the foregoing polymers and a polyolefin elastomer, an
5 alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers, and the print skin layer (B) comprises a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an
10 alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers.

Figure 1 illustrates a polymeric film of the present invention in which relatively thin skin layers (B) and (C) respectively overlie and underlie a relatively thick core layer (A).

Adhesive Label Stock and Adhesive Label Construction

The polymeric film of the present invention comprises a multilayered film that is used with an adhesive layer to label an article. The article can be any shaped container to include a bottle, a jug, a jar or a drum. The article can
20 be made of paper, cardboard, glass, metal, plastic, or a mixture thereof. The article can be any shaped container used for packaging a consumer or industrial product. In an embodiment of the invention the film comprising layers (A), (B) and (C) further comprises an adhesive layer (D) having an upper and a lower surface wherein the upper surface of the adhesive layer is
25 adhesively joined to the lower surface of the adhesive skin layer (C). In another embodiment of the invention the adhesive layer comprises an adhesive as described hereinbelow. The polymeric film is generally converted to an adhesive containing label stock and then to an adhesive label which is used to label an article. The polymeric film can be converted to an adhesive
30 containing label stock by initially slitting the film into widths suitable for labeling articles. The slit film is then combined with an adhesive layer and a release liner to form an adhesive containing label stock as illustrated in Figure 2. The adhesive layer can be combined with the release liner initially, the slit polymeric film initially, or with both liner and film simultaneously. In Figure 2,

an adhesive layer (D) has an upper and a lower surface where the upper surface of the adhesive layer (D) is adhesively joined to the lower surface of the adhesive skin layer (C) of the polymeric film comprising layers (A), (B) and (C), and a release liner (E) is releasably adhered to the lower surface of the adhesive layer (D). The release liner can comprise a monolayered or multilayered film where the film of the liner can comprise a paper film, a polymeric film such as for example a polyolefin film, or a mixture of any of the foregoing films. The release liner normally comprises a release coating on the film of the release liner such as for example an organosiloxane polymer, which is also known as a silicone, so that the release liner can later be removed from the adhesive layer during the labeling process. The adhesive layer comprises an adhesive that comprises one or more thermoplastic polymers to include one or more elastomeric polymers. In an embodiment of the invention, the adhesive layer comprises a pressure sensitive adhesive. In embodiments of the invention the adhesive layer comprises a pressure sensitive adhesive which comprises a solvent adhesive, an emulsion adhesive, or a hot melt adhesive.

Pressure sensitive hot melt adhesives generally comprise elastomeric polymers which comprise natural rubbers and/or synthetic polymers and additional components such as for example tackifiers as described in US Patent No. 6214935. The hot melt adhesive comprises an alkene-unsaturated carboxylic acid ester copolymer such as for example ethylene-vinyl acetate copolymers and ethylene-ethyl acrylate copolymers, a styrene polymer and/or copolymer and/or block copolymer such as, for example, styrene-isoprene-styrene block copolymers and styrene-butadiene-styrene block copolymers, a polyolefin, a polyamide, a polyester, a polyurethane such as for example reactive polyurethanes, and a mixture of any of the foregoing polymers. The hot melt adhesive further comprises tackifiers, plasticizers, waxes, performance additives including antioxidants and photoinitiators and fillers and reinforcing agents and compatibilizers and nucleating agents and mixtures of any of the foregoing additives, and mixtures of any of the foregoing components. In an embodiment of the invention, the adhesive layer comprises a hot melt adhesive which comprises a styrene block copolymer.

The adhesive containing label stock is typically converted to an adhesive label by initially printing the label stock using any printing means such as, for example, offset lithography with a suitable printing ink to include for example water-based inks and solvent-based inks and UV-activated inks.

5 Alternatively, the slit film can be printed prior to combining with the hot melt adhesive and release liner. The printed label stock is then die-cut into a series of individual adhesive labels that can be used to label articles of commerce in a labeling operation. In the labeling operation, a series of die-cut adhesive labels can be peeled away one by one from a matrix of the polymeric film
10 bordering the die-cut label and the underlying release liner.

Examples

The following nonlimiting examples further describe and illustrate the present invention.

15

Example 1: A 2.5 mil (63.5 micrometers) film is prepared by coextrusion, orientation by stretching in the machine direction 5.3 times, annealing, and corona discharge treatment of the print skin layer. Core layer (A) is 92% of film thickness and comprises on a wt. basis 100% polypropylene
20 copolymer. Print skin layer (B) is 4% of film thickness and comprises on a wt. basis 88% polypropylene copolymer, 10% polyethylene plastomer, and 2% antiblock agent. Adhesive skin layer (C) is 4% of film thickness and comprises on a wt. basis 98% polypropylene homopolymer and 2% antiblock agent.

25

Example 2: A 2 mil (50.8 micrometers) film is prepared as in Example 1. Core layer (A) is 80% of film thickness and comprises on wt. basis 90% polypropylene homopolymer and 10% polyethylene plastomer. Print skin layer (B) is 9% of film thickness and comprises on wt. basis 89%
30 polypropylene homopolymer, 10% polyethylene plastomer and 1% antiblock agent. Adhesive skin layer (C) is 11% of film thickness and comprises on wt. basis 98% polypropylene homopolymer and 2% antiblock agent.

Example 3: A 2.5 mil (63.5 micrometers) film is prepared as in Example 1. Core layer (A) is 74% of film thickness and comprises on wt. basis 90% polypropylene copolymer and 10% polyethylene plastomer. Print skin layer (B) is 11% of film thickness and comprises on wt. basis 61% polypropylene copolymer, 36% polyethylene elastomer and 3% antiblock agent. Adhesive skin layer (C) is 15% of film thickness and comprises on wt. basis 98% polypropylene homopolymer and 2% antiblock agent.

Example 4: A 2 mil (50.8 micrometers) film is prepared as in Example 1. Core layer (A) is 80% of film thickness and comprises on wt. basis 90% polypropylene copolymer and 10% polyethylene plastomer. Print skin layer (B) is 9% of film thickness and comprises on wt. basis 89% polypropylene copolymer, 10% polyethylene plastomer, and 1% antiblock agent. Adhesive skin layer (C) is 11% of film thickness and comprises on wt. basis 98% polypropylene homopolymer and 2% antiblock agent.

Example 5: A 2.5 mil (63.5 micrometers) film is prepared as in Example 1. Core layer (A) is 92% of film thickness and comprises on wt. basis 100% polypropylene copolymer. Print skin layer (B) is 4% of film thickness and comprises on wt. basis 88% polypropylene copolymer, 10% polyethylene plastomer, and 2% antiblock agent. Adhesive skin layer (C) is 4% of film thickness and comprises on wt. basis 98% polypropylene homopolymer and 2% antiblock agent.

Example 6: A 2 mil (50.8 micrometers) film is prepared as in Example 1. Core layer (A) is 74% of film thickness and comprises on wt. basis 90% polypropylene copolymer and 10% polyethylene plastomer. Print skin layer (B) is 11% of film thickness and comprises on wt. basis 61% polypropylene homopolymer, 36% polyethylene plastomer, and 3% antiblock agent. Adhesive skin layer (C) is 15% of film thickness and comprises on wt. basis 83% polypropylene homopolymer, 15% polypropylene copolymer and 2% antiblock agent.

Example 7: A 2 mil (50.8 micrometers) film is prepared as in Example 1. Core layer (A) is 78% of film thickness and comprises on wt. basis 90% polypropylene copolymer and 10% polyethylene plastomer. Print skin layer (B) is 12% of film thickness and comprises on wt. basis 88% polypropylene copolymer, 10% polyethylene plastomer, and 2% antiblock agent. Adhesive skin layer (C) is 10% of film thickness and comprises on wt. basis 78% polypropylene homopolymer, 20% polypropylene copolymer and 2% antiblock agent.

Example 8: A 2.5 mil (63.5 micrometers) film is prepared as in Example 1. Core layer (A) is 77% of film thickness and comprises on wt. basis 100% polypropylene homopolymer. Print skin layer (B) is 15% of film thickness and comprises on wt. basis 88% polypropylene copolymer, 10% linear low density polyethylene, and 2% antiblock agent. Adhesive skin layer (C) is 8% of film thickness and comprises on wt. basis 93% polypropylene homopolymer, 5% polypropylene copolymer and 2% antiblock agent.

Examples 9-20 are monolayered adhesive skin films that are prepared on a pilot line by extrusion, orientation by stretching 5 times in the machine direction, and annealing. The films of Examples 9-20 are evaluated for hot melt adhesive performance in terms of resistance to swelling and distortion by using mineral oil, which mimics additive migration of hot melt adhesives. Table I lists the hot melt adhesive performance for the films of Examples 9-20.

Table I

Example No.	Composition ^a	Rating @ 16 Hours ^b	Rating @ 42 Hours ^b
9	80% RCP ¹ + 20% HPP ¹	1	4
10	50% RCP ¹ + 50% HPP ¹	1	2
11	20% RCP ¹ + 80% HPP ¹	1	1
12	100% HPP ¹	1	1
13	100% RCP ¹	1	4
14	50% GMAH ¹ + 48% RCP ² + 2% AB	5	5
15	98% GMAH ¹ + 2% AB	5	5
16	68% RCP ² + 30% PT + 2% AB	1	5
17	77% GMAH ¹ + 21% HPP ² + 2% AB	1	4
18	40% GMAH ² + 55% HPP ² + 5% AB	2	4
19	15% GMAH ³ + 80% HPP ² + 5% AB	1	3
20	95% GAA + 5% AB	2	3

^a Composition is on a weight basis.

RCP¹ is Basell XL451J polypropylene random copolymer having melt flow rate (MFR) of 3.9.

HPP¹ is Basell SR257M polypropylene homopolymer having MFR of 2.0.

- 5 GMAH¹ is Mitsui Admer QF551A polypropylene homopolymer grafted with 0.5% maleic anhydride and having MFR of 5.7.

RCP² is Huntsman P5M4K-072X nucleated polypropylene random copolymer having MFR of 10.0.

- 10 AB is Ampacet 401960 antiblock agent which contains 3% poly(methyl methacrylate) in polypropylene homopolymer carrier and has MFR of 7.2.

PT is ExxonMobil Exxelor PA609A polyterpene rosin in polypropylene homopolymer carrier.

HPP² is Huntsman P4C4K-173X nucleated polypropylene homopolymer having MFR of 12.0.

GMAH² is Chemtura Polybond 3150 polypropylene homopolymer grafted with 0.5% maleic anhydride and having MFR of 50.0.

- 15 GMAH³ is Chemtura Polybond 3200 polypropylene homopolymer grafted with 1.0% maleic anhydride and having MFR of 115.

GAA is Chemtura Polybond 1001 polypropylene homopolymer grafted with 6% acrylic acid and having a MFR of 40.0.

^b The rating scale for film distortion due to additive migration using mineral oil is as follows:

- 20 1 = flat (no discernable warpage),
 2 = low warpage,
 3 = medium warpage,
 4 = high warpage,
 5 = curled.

25

Each of the documents referred to in the Detailed Description is incorporated herein by reference. All numerical quantities in this application used in the Detailed Description and appended Claims are understood to be

modified by the word "about" except in examples and where explicitly indicated otherwise. All range and ratio limits in the Detailed Description and appended Claims are combinable.

5 While the invention has been presented in the Detailed Description and appended Claims, it is understood that various modifications of this invention will become apparent to those skilled in the art upon reading this application. Therefore these various modifications, that fall within the scope of the appended Claims and/or Detailed Description, are considered to be a part of this invention.

10

What is claimed is:

1. A polymeric film for labeling articles, comprising:

5 (A) a core layer having an upper surface and a lower surface and comprising a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers;

10 (B) a print skin layer comprising a polypropylene homopolymer, a polypropylene copolymer, a high density polyethylene, or a mixture of any of the foregoing polymers and at least 10% by weight based on the weight of the layer (B) of a polyolefin elastomer, an alkene-unsaturated carboxylic acid or acid derivative copolymer, or a mixture of any of the foregoing polymers wherein the print skin layer overlies the upper surface of the core layer; and

15 (C) an adhesive skin layer comprising a polypropylene homopolymer wherein the adhesive skin layer has an upper surface and a lower surface wherein the upper surface of the adhesive skin layer underlies the lower surface of the core layer;

wherein the composition of layer (C) is different from the composition of layer (A), and the polymeric film is oriented.

20 2. The film of claim 1 wherein the polypropylene homopolymer of layer (C) is an isotactic polypropylene homopolymer having an isotactic stereoregularity greater than 90%.

25 3. The film of claim 1 wherein the polypropylene homopolymer of layer (C) is a syndiotactic polypropylene homopolymer having a syndiotactic stereoregularity greater than 90%.

30 4. The film of claim 1 wherein the polypropylene homopolymer of layer (C) is a mixture of an isotactic polypropylene homopolymer and a syndiotactic polypropylene homopolymer.

5. The film of any one of the preceding claims wherein the layer (C) further comprises a nucleating agent, an antiblock agent, a processing

aid, a slip agent, an antistatic agent, an antioxidant or a mixture of any of the foregoing additives.

6. The film of any one of the preceding claims wherein layer (A) or
5 layers (A) and (B) comprise a nucleating agent.

7. The film of any one of the preceding claims wherein layer (A)
further comprises a polyolefin elastomer, an alkene-unsaturated carboxylic
acid or acid derivative copolymer, or a mixture of any of the foregoing
10 polymers.

8. The film of claim 7 wherein the polyolefin elastomer comprises a
medium density polyethylene, a linear low density polyethylene, a very low
density polyethylene, a low density polyethylene prepared by free radical
15 catalysis, a polyethylene plastomer, a polypropylene copolymer elastomer, or
a mixture of any one of the foregoing polymers.

9. The film of claim 7 wherein the alkene-unsaturated carboxylic
acid or acid derivative copolymer comprises an alkene-unsaturated carboxylic
20 acid copolymer, an alkene-unsaturated carboxylate ester copolymer, a metal
salt of an alkene-unsaturated carboxylic acid copolymer, or a mixture of any of
the foregoing copolymers.

10. The film of any one of the preceding claims wherein the
25 thickness of layer (C) is 1% to 30% of the thickness of the polymeric film.

11. The film of any one of the preceding claims wherein the film is
oriented by stretching the film from 2 to 12 times in the machine direction or in
both the machine and transverse directions.

30

12. An adhesive containing label stock for use in adhesive labels,
comprising:

the film of any one of the preceding claims;

(D) an adhesive layer having an upper and a lower surface wherein the upper surface of the adhesive layer is adhesively joined to the lower surface of layer (C); and

(E) a release liner wherein the release liner is releasably
5 adhered to the lower surface of the adhesive layer (D).

13. The adhesive containing label stock of claim 12 wherein the adhesive layer comprises a hot melt adhesive.

10 14. An adhesive label die-cut from the label stock of any one of claims 12 or 13.

(B)
(A)
(C)

Figure 1

(B)
(A)
(C)
(D)
(E)

Figure 2