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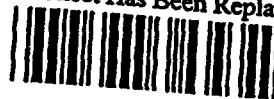
OPAQUE ORIENTED POLYMERIC FILM
STRUCTURE AND METHOD OF PREPARING SAME

This invention relates to polymer films of enhanced opacity; and to methods of making such films. More particularly, the invention relates to a biaxially oriented composite such film structures; and to their preparation by coextrusion.

U. S. Patent No. 4,377,616 discloses an opaque, biaxially oriented polymer film structure comprising a thermoplastic polymer matrix film core layer possessing numerous voids, a substantial number of which contain at least one spherical void-initiating particle, and transparent, void-free thermoplastic skin layers adhering to the surface of the core layer. The unique structure of the core layer imparts a much higher degree of opacity, possibly due to the effects of light scattering, than that possible from the use of an opacifying pigment alone. Transparent, void-free thermoplastic skin layers are adhered to the surface of the core layer of the film of U. S. Patent No. 4,377,616 to yield smooth film surfaces, free of the surface irregularities or surface projections of the core material. Without such skin layers being provided, U. S. Patent No. 4,377,616, teaches that the lustrous appearance of the satin finish would be materially lessened.

However, the use of transparent, void-free thermoplastic skin layers to encapsulate the core layer can yield a film with somewhat undesirable end use characteristics. For example, when such films are utilized in certain label making operations, a skin layer surface may, through frictional contact, adhere to an excessive degree to the label processing equipment, resulting in labels of poor quality and/or equipment shut-down. Although the equipment can often be adjusted to minimize such problems, films which would provide a greater degree of operability would clearly be desirable.

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Another problem sometimes encountered in the end-use processing of the known opaque multilayer film structures employing transparent, void-free thermoplastic skin layers is that often such film structures are difficult to cut. Additionally, from an appearance standpoint, rather than provide a transparent skin layer, it is sometimes desirable to provide a skin layer of increased opacity.

This invention seeks to provide a film structure of high opacity which provides improved cutting characteristics and an improved range of operability when undergoing processing, while maintaining high strength and stiffness.

In accordance with one aspect of this invention, there is provided an opaque, biaxially oriented polymeric film structure comprising:

(a) a core layer, having a first surface and a second surface, comprising a thermoplastic polymeric matrix within which is located a stratum of voids, a substantial number of the voids containing at least one spherical, void-initiating particle which is phase distinct and incompatible with the matrix material, the population of voids in the core being such as to cause a significant degree of opacity; and

(b) at least one skin layer, adhering to at least the first surface of the core layer, comprising a thermoplastic polymeric matrix within which is located a stratum of voids, a substantial number of said voids containing at least one void-initiating solid particle which is phase distinct and incompatible with the matrix material.

In accordance with a further aspect of this invention, there is provided a process for preparing an opaque, biaxially oriented polymeric film structure, comprising the steps of:

(a) mixing a major proportion of a first thermoplastic polymeric material with a minor proportion of a first material of

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higher melting point, or having a higher glass transition temperature, than the first thermoplastic polymeric material to produce a core layer mixture;

(b) heating the core layer mixture produced in step (a) to a temperature at least above the melting point of the first thermoplastic polymeric material;

(c) dispersing the first material uniformly throughout the molten first thermoplastic polymeric material in the form of spheres;

(d) mixing a major proportion of a second thermoplastic polymeric material with a minor proportion of a second material of higher melting point, or having a higher glass transition temperature than the second thermoplastic polymeric material to produce a skin layer mixture;

(e) heating the skin layer mixture produced in step (d) to a temperature least above the melting point of the second thermoplastic polymeric material;

(f) dispersing the second material uniformly throughout the molten second thermoplastic polymeric material;

(g) forming an opaque, biaxially oriented polymeric film structure from the core layer mixture and the skin layer mixture, the forming step comprising coextrusion conducted at a temperature and to a degree to form strata of opacifying voids within both the core layer and the void-containing skin layer.

In order to achieve the desired film strength, stiffness and opaque appearance of the multilayer film structure of the present invention, it is desirable that a particular thickness relationship exist between the thickness of the core layer, the thickness or thicknesses of the void-containing skin layer or layers adhered to one or both surfaces of the core layer, and the thickness of the optional void-free skin layer which can be transparent or pigmented for further opaqueness. It is preferred

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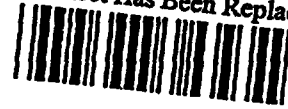
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that the core layer thickness be from 30% to 95% of the overall thickness of the structure. This, in combination with the population and configuration of the voids in a structure of at least 0.5 mils thick, will materially contribute to the overall degree of opacity of the structure. The population and configuration of the voids and the core thickness will be such as to cause a significant degree of opacity, for example, less than 50% light transmission, with less than 70% light transmission preferred. Similarly, by maintaining the skin thickness within a particular range in relation to the thickness of the core layer, the overall combination can result in an opaque lustrous appearance.

Other structures besides the three layer structure described above are within the scope of the present invention. For example, structures having more than three layers and having a plurality of void-containing layers would have particular utility in certain applications. Such a film could employ different void-initiating agents of the types described hereinbelow in each of the void-containing layers and/or have each of its void-containing layers voided to a different extent via processing, size of the void-initiating particles and/or number of particles employed. As can be appreciated, both the physical properties of the resultant multilayered film, as well as its visual appearance can be advantageously varied using this embodiment of the film approach of the present invention.

To prepare the core layer of the film structure of the present invention, it is preferred that the average diameter of the void-initiating particles used in that layer be from 0.1 to 10 microns. These particles should be generally spherical in shape so as to initiate a void of unusual regularity and orientation in a stratified relationship throughout the thermoplastic polymer

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matrix material after biaxial orientation of the structure. This does not mean that every void is the same size; rather, what is meant is that, in general, each void tends to be of like shape even though it may vary in its dimensions from those of other voids because all of the voids are initiated by spherical particles. Ideally, the voids assume a shape defined by two opposed and edge-contacting concave disks. Optimum characteristics of opacity and satin-like appearance are obtained when the two average major void dimensions are greater than about 30 microns. The void-initiating particle material for use in the thermoplastic polymer matrix core layer should be incompatible with the substrate material, at least at the temperature of biaxial orientation.

The core layer has been described above as comprising a thermoplastic polymer matrix material within which is located a stratum of voids. The term "stratum" is intended to convey the understanding that there are a large number of voids within matrix and the voids themselves are oriented so that the two major dimensions are aligned in correspondence with the direction of orientation of the polymeric film structure. The opacity of the system may be further enhanced by the inclusion of an opacifying pigment; for example in an amount from 1% to 3% by weight dispersed throughout the core layer. The pigment material can be present in such a particle size and shape that it does not, at least in any material sense, contribute any void initiation by itself. The optional presence of the opacifying pigment may contribute 3% to 8% to the degree of opacity of the system.

The thermoplastic polymeric material comprising the matrix of the core layer and the void-initiating particle must be incompatible by forming two distinct phases. The spherical void-initiating particles constitute a dispersed phase throughout

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the lower melting thermoplastic polymeric matrix which will, upon orientation, become a void-filled matrix with the spherical particles contained somewhere within the voids.

The biaxial orientation to form the film structure of this invention described herein, in addition to rendering its core layer opaque, improves other physical properties of the multilayer structure such as flex-cracking resistance, Elmendorff tear strength, elongation, tensile strength, impact strength and cold strength properties. In addition to this, an unusual and highly attractive lustrous appearance is imparted to the film. This appearance is generally not seen in the absence of the skin layer or layers, or if the thickness of the skin layer or layers is not enough to conceal the surface imperfections produced within the core layer. The resulting film can also possess low water vapor transmission rate and low oxygen transmission rate characteristics which makes the film ideally suited for packaging food products, including liquids. The film also has attractive utility as a decorative wrap material. Generally, a greater number of voids will be closed, as a result of which there is essentially no opportunity for liquid and/or gas to penetrate the core layer.

The void-initiating particles can be organic or inorganic provided that they are approximately spherical in shape and within the preferred particle size range so as to initiate the formation of a void, which, in a significant number of cases, will take on a lens-like shape, that is, a lens of a biconvex shape. When a polymeric material is employed as the void-initiating particle, it can be a polymer which is co-melted with the polymer of the core layer. In such a case, it is necessary for the void-initiating polymer to have a sufficiently higher melting point than the core polymer to be incompatible therewith and to be capable of assuming a dispersed phase in the form of small spherical particles as the temperature of the melt is reduced. It is preferred

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that the void initiating particles can be preformed and then uniformly dispersed into a melt of core polymer, for example, one of polypropylene, thereby minimizing the risk of thermal degradation of the core thermoplastic polymeric matrix material.

It is believed that, because of the number, shape and orientation strata-wise of the voids in the core layer, a significantly enhanced light scattering effect is obtained. This effect is further increased by the presence of a void-free transparent or pigmented skin layer coextensively applied to the core layer.

When preformed void-initiating particles are employed, it is the shape and size of the particles that is important rather than their chemical nature. Thus, solid or hollow organic or inorganic particles of any type can be employed. Interesting effects can be produced by the use of spheres of different colors. Since, statistically, each void has approximately one particle somewhere within the void, interesting and aesthetically pleasing color and/or reflectance effects can be imparted to the overall layer structure by use of particles of different color absorption or reflectance. The light scattered in a particular void is additionally either absorbed or reflected by the void-initiating particle and a separate color contribution is made to the light scattered within each void.

The void-initiating particles can be based on any of a variety of thermoplastic resins; for example, polybutylene terephthalate, polyethylene terephthalate acetals, polyamides, or acrylic resins, provided that they remain phase distinct and incompatible with the resin constituting the remainder of the core layer. Polybutylene terephthalate (PBT), also known as polytetramethylene terephthalate (PTMT), is an especially preferred void-initiating material for use in the present invention, typically involving processing from 450°-500°F.

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Inorganic materials which are useful as void-initiating particles for the core matrix layer include solid or hollow preformed glass spheres, metal beads or spheres and ceramic spheres. In fact any material which is inherently spherical, or can be formed into a sphere without causing thermal degradation to the core polymer, is entirely suitable herein.

The incorporation of voids in the core layer of the film structures of the invention permits a reduction in light transmission therethrough to as low as 16% and even lower. This would be the case for a film having an overall thickness of at least 1.5 mils when the core portion is at least 60% and the thickness of the skin layer or layers makes up the remaining 40% thickness.

While the preferred particle size of the void-initiating particles is from 0.1 to 10 microns, it is particularly preferred that the particle size is from 0.75 to 2 microns. The void-initiating particles can be present in up to 20% by weight of the core layer prior to orientation, a preferred range being from 2 to 7 percent by weight.

Suitably, the film structure of this invention may further comprise a void-free thermoplastic skin layer adhering to the second surface of the core layer, the void-free skin layer being of a thickness such that the asperities of the core layer are not, at least substantially, manifest. Such a void-free skin layer may contain an opacifying agent, a coloring agent or an antiblocking agent.

However has now been discovered that, by incorporating voids into at least one of the skin layers (b) through the use of inorganic void-initiating particles, the coefficient of friction of the surface of that skin layer can be significantly reduced, thus extending the range of operability of such a film in film processing equipment. Suitably, a second skin layer (b) is also

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adhered to the second surface of the core layer. Moreover, by employing the void-initiating particles of the type preferred for use in the practice of the present invention, the problems associated with dislodged particles, is avoided. Although this benefit may suggest the use of certain preferred inorganic void-initiating particles in a non-encapsulated, single layer structure, to achieve the same level of opaqueness as is obtained using the organic void-initiating particles preferred for use in the core layer, higher concentrations of inorganic material are required. These higher concentrations would serve to sacrifice many of the desirable attributes inherent in the polymeric matrix material, itself, such as strength and stiffness.

Another advantage which accrues from the use of at least one void-containing skin layer in the multilayer film structures of the present invention relates to an improvement in film cutting. Surprisingly, films produced according to the present invention have been found to cut in a more paper-like manner, which is advantageous when such films are used in the production of label stock for use in various packaging applications, such as bottle labels. The use of a void-containing skin layer on the side of the film where the final cut takes place creates a weaker region of film in this critical area, while retaining overall film integrity. This weaker region of film is responsible for the more paper-like cutting characteristics of the films produced in accordance with the present invention. Such paper-like cutting characteristics have been found to particularly benefit the production of coextruded pressure sensitive label stock material having a peelable backing affixed thereto.

Void-initiating particles preferred for use in at least one of the skin layers of the multilayer structure of the present invention include both inorganic and organic materials. When an organic material is employed as a void-initiating particle, it is

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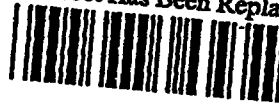
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important that the material be one which does not exhibit the tendency of becoming easily dislodged from the matrix structure, as discussed above. Preferred inorganic materials include crushed calcium carbonate, magnesium carbonate, magnesium silicate, aluminum silicate, silica and alumina, with calcium carbonate being particularly preferred. As was the case for the void-initiating particles preferred for use in the core layer, the preferred particle size of the void-initiating particles is from 0.1 to 10 microns, while it is particularly preferred that the particle size range from 1 to 5 microns. The void-initiating particles can be present in an amount up to 70% by weight of the skin layer prior to orientation, a preferred range being from 5 to 20 percent by weight. Compared to the particles preferred for use in the core layer, these particles, and particularly the inorganic particles, generally have a random shape, rather than a spherical shape, the shape of the particles used in the skin layer not being critical to the appearance of the resulting multilayer structure. Consistent with the void-initiating particles used in the core layer, those particles selected for use in at least one of the skin layers must be incompatible with the polymeric matrix skin layer material.

Polymers preferred for use in the practice of the present invention are isotactic propylene homopolymers, random and sequential copolymers of propylene and ethylene, and other copolymers composed primarily of propylene units with isotactic polypropylene being preferred for the core layer and for the skin layer. Such polymeric materials will generally exhibit melting points of at least 285°F., preferably of at least 300°F. Melt indices in the range of 0.5 g/10 minutes to about 8 g/10 minutes at 446°F. are preferred.

Additives such as fillers, antioxidants, pigments, antistatic agents, slip agents and antitack agents can be incorporated in the usual amounts in one or more of the layers constituting the multilayer film.

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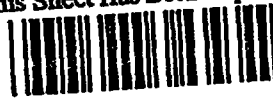
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As disclosed in U. S. Patent No. 4,701,370 the addition of a relatively minor amount of particles of titanium dioxide-coated mica to the polymeric material constituting the skin layer or layers provides a marked increase in the stiffness of the multilayer film. An effective amount of titanium dioxide-coated mica for use in the practice of the present invention would be from 0.1 to 10 weight percent of the skin layer material. A suitable titanium dioxide-coated mica is the Eagle Mica M series marketed by Eagle Quality Products Co., of Uwckland, PA.

It is preferred to coextrude the core layer and the skin layer or layers in producing the multilayer film of this invention. Thereafter, the film, whether produced by coextrusion or by some other technique is subjected to biaxial stretching, or orientation, to an extent and at a temperature calculated to provide the maximum degree of opacity without any significant impairment in the physical characteristics, including the appearance of the film. Obviously, as the materials employed in the construction of the film differ, the conditions of biaxial orientation will be modified to reflect such differences. By way of exemplification, when employing a stereoregular polypropylene as the core polymer and when employing PBT as the void-initiating material, a machine direction orientation from 4 to 8 times and a transverse direction orientation from 4 to 8 times, at a drawing temperature from 212°F. to 320°F. can be used to provide an opaque, biaxially oriented film from 0.5 to 2 mils in overall thickness.

When producing coextruded pressure sensitive label stock material with peelable backing, it is advantageous from the standpoint of label cutting to position the void-containing skin layer against the peelable backing material. The pressure sensitive adhesive can be applied to the surface of the void-containing skin layer in a continuous or discontinuous manner as dictated by the requirements of the particular application. The adhesive can be applied using any of a number of well-known techniques for

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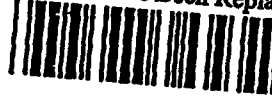
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accomplishing such tasks, including the technique of "printing" the adhesive upon the exposed surface of the void-containing skin layer.

The pressure sensitive adhesive component can be selected from amongst any of the materials which are known to be useful for this purpose. In general, non-solvent pressure sensitive adhesive materials are preferred to solvent-based pressure sensitive adhesive materials. Acceptable materials include: natural and/or synthetic rubbers, butadiene-styrene rubbers, polyisobutylene rubbers, polyisobutadiene rubbers, ethylene-propylene terpolymer rubbers, polyacrylic rubbers, polyvinyl ether rubbers, butadiene-acrylonitrile rubbers, butadiene-styrene-acrylonitrile rubbers or butyl rubbers, in combination with a tacky or tackifying resin, such as a rosin ester, a hydrogenated rosin ester, the glycerol ester of hydrogenated rosin, a polyterpene resin, a crude rubber, a polyisobutylene or other unsaturated hydrocarbon resin, a phenolated terpene or a petroleum resin. The pressure sensitive adhesive may also contain additives such as antioxidants or fillers.

The polymeric release layer employs a release agent applied to the film surface to which it will subsequently be affixed to the pressure sensitive adhesive side of the multilayer opaque oriented film structure. The release agent can be selected from any of the materials known to have been used for their release properties for pressure sensitive adhesives. Common types are silicones and modified silicones, the modification including both copolymerization, or silicones with other non-release chemical agents, or by adding non-silicone materials to a silicone coating prior to surface application of the release agent to the release layer. Other release agents such as polyethylene, fluorocarbon, the Werner-type chromium complexes and polyvinylidene octadecyl carbamate have also been used. The choice of a release coating is dependent on the tack and adhesion level of the adhesive contained

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in the adjacent layer as may be appreciated by those skilled in the art. Any of the well-known techniques for applying release agents may be employed herein.

If desired, the exposed surface of one of the skin layers can be treated in a known and conventional manner, such as by flame or corona discharge to achieve the expected benefits obtained by such surface treatments, such as improved ink receptivity.

The following Examples are illustrate the invention:

EXAMPLE 1

A mixture of 94 percent, by weight, isotactic polypropylene (MP = 320°F., melt index = 4.5), containing 6 weight percent PBT (MP = 440°F.) as the core layer void-initiating material, is melted in an extruder with a screw of L/D ratio of 20/1 to provide the core layer mixture. A second extruder, in association with the first extruder, is supplied with the same isotactic polypropylene as the first extruder but containing titanium dioxide particles at 4 percent, by weight, and talc (Cyprus Mistron ZSC) at 2 percent, by weight. The talc serves as an antiblocking agent, while the titanium dioxide particles are employed as a whitener for this skin layer mixture. A third extruder, in association with the first two extruders, is supplied with the same isotactic polypropylene as the first two extruders but containing 15 percent, by weight, of calcium carbonate in crushed, random particle form, to serve as void-initiating particles for this skin layer mixture. A melt coextrusion is carried out while maintaining the cylinder of the core polymer material at a temperature sufficient to melt the polymer mixture, i.e., from about 450°F. to about 550°F. or higher. The polypropylene mixtures to be extruded as skin layers are maintained at about the same temperature as the polypropylene

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used in fabricating the core layer. A multilayer film laminate is coextruded with a core thickness representing about 70 percent of the overall extruded thickness, with the thicknesses of the skin layers representing the balance of the film thickness. The unoriented film measured about 40 mils in thickness. The resultant film sheet was subsequently oriented eight by five and one-half times using a commercially available sequential biaxially orienting apparatus to provide a multilayer film structure. The machine direction (MD) orientation is conducted at about 285°F. and the transverse direction (TD) orientation is conducted at about 300°F. The resultant 1.5 mil multilayer film exhibits a relatively rough surface finish of high opacity when viewed from its void-containing skin layer side and a smooth lustrous appearance when viewed from its non-void containing skin layer side.

The film so produced was compared to a film laminate produced in accordance with U. S. Patent No. 4,560,614, the contents of which are incorporated by reference in their entirety. The film produced in Example 1 exhibited a wider range of operability in a beverage container label making apparatus than the film produced in accordance with U. S. Patent No. 4,560,614. This was believed to result from the reduced coefficient of friction of the void-containing skin layer surface. Moreover, from an appearance standpoint, the film produced in Example 1 exhibited enhanced opacity as a result of the void-containing skin layer utilized.

EXAMPLE 2

This example pertains to the production of pressure sensitive label stock. Once again, a mixture of 94 weight percent isotactic polypropylene (MP = 320°F., melt index = 4.5), containing 6 weight percent PBT (MP = 440°F.) as the core layer

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void-initiating material, is melted in an extruder with a screw of L/D ratio of 20/1 to provide the core layer mixture. A second extruder, in association with the first extruder is supplied with the same isotactic polypropylene as the first extruder and titanium dioxide particles at 4 percent, by weight, and talc (Cyprus Mistron ZSC) at 2 percent, by weight. A third extruder in association with the first two extruders, is supplied with the same isotactic polypropylene as the first two extruders and 15 percent, by weight, of crushed calcium carbonate, to serve as void-initiating particles for this layer. A melt coextrusion is carried out while maintaining the cylinder of the core polymer material at a temperature sufficient to melt the polymer mixture, i.e., from about 450°F. to about 550°F., or higher. As before, the polypropylene mixtures coextruded as skin layers are maintained at about the same temperature as the polypropylene used in fabricating the core layer. A multilayer film laminate is coextruded with a core thickness representing about 70 percent of the overall extruded thickness, with the thicknesses of the skin layers representing the balance of the film thickness. The unoriented film measured about 40 mils in thickness. The resultant film is biaxially oriented as in Example 1. A 2:1 weight-part mixture of a hydrogenated rosin derivative and styrene-butadiene rubber pressure-sensitive adhesive is printed onto the void-containing skin layer surface. A release layer, also employing the same polypropylene as used in the multilayer coextruded film structure, is separately extruded. A commercially obtained silicone release agent is applied to one surface of the release layer. The multilayer opaque film is adhered to the separately extruded, transparent release layer.

The pressure sensitive label stock so produced was run through a die-cutting apparatus to produce label stock having separately detachable labels affixed to a continuous release layer.

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When compared to label stock produced in accordance with with U. S. Patent No. 4,582,736, the contents of which are incorporated by reference in their entirety, the film produced in accordance with the present invention exhibited improved cutting characteristics over the pressure sensitive label stock produced in accordance with U. S. Patent No. 4,582,736. This was believed to result from the void-containing layer surface. Moreover, from an appearance standpoint, the film produced in Example 2 exhibited enhanced opacity as a result of the void-containing skin layer utilized.

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CLAIMS FOR AUSTRALIA, FINLAND, JAPAN, N.Z. NORWAY, EPC

1. An opaque, biaxially oriented polymeric film structure comprising:

(a) a core layer, having a first surface and a second surface, comprising a thermoplastic polymeric matrix within which is located a stratum of voids; a substantial number of the voids containing at least one spherical, void-initiating particle which is phase distinct and incompatible with matrix material, the population of voids in the core being such as to cause a significant degree of opacity; and

(b) at least one skin layer, adhering to at least the first surface of the core layer, comprising a thermoplastic polymeric matrix within which is located a stratum of voids, a substantial number of said voids containing at least one void-initiating solid particle which is phase distinct and incompatible with the matrix material.

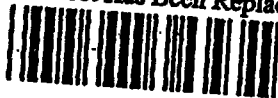
2. A film structure according to claim 1, which further comprises a void-free, thermoplastic skin layer adhering to the second surface of the core layer, the void-free skin layer being of a thickness such that the asperities of the core layer are not, at least substantially, manifest.

3. A film structure according to claim 2, wherein the void-free skin layer contains an opacifying agent, a coloring agent, or an antiblocking agent.

4. A film structure according to claim 1, wherein a second skin layer (b) is also adhered to the second surface of the core layer.

5. A film structure according to claim 4, wherein the second skin layer (b) is voided to an extent which differs from that of the first skin layer (b), as indicated by relative opacity.

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6. A film structure according to claim 4 or 5, wherein the void-initiating solid particles of the second skin layer (b) comprises a material different from that of the void-initiating solid particles of the first skin layer (b).

7. A film structure according to any preceding claim, wherein the core layer comprises isotactic polypropylene.

8. The film structure according to any preceding claim, wherein the skin layer comprises from isotactic polypropylene.

9. A process for preparing an opaque, biaxially oriented polymeric film structure, comprising the steps of:

(a) mixing a major proportion of a first thermoplastic polymeric material with a minor proportion of a first material of higher melting point, or having a higher glass transition temperature, than the first thermoplastic polymeric material to produce a core layer mixture;

(b) heating the core layer mixture produced in step (a) to a temperature at least above the melting point of the first thermoplastic polymeric material;

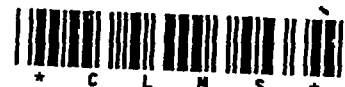
(c) dispersing the first material uniformly throughout the molten first thermoplastic polymeric material in the form of spheres;

(d) mixing a major proportion of a second thermoplastic polymeric material with a minor proportion of a second material of higher melting point, or having a higher glass transition temperature than the second thermoplastic polymeric material to produce a skin layer mixture;

(e) heating the skin layer mixture produced in step (d) to a temperature least above the melting point of the second thermoplastic polymeric material;

(f) dispersing the second material uniformly throughout the molten second thermoplastic polymeric material;

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(g) forming an opaque, biaxially oriented polymeric film structure from the core layer mixture and the skin layer mixture, said forming step comprising coextrusion conducted at a temperature and to a degree to form strata of opacifying voids within both the core layer and the void-containing skin layer.

10. A process according to claim 9 which further comprises the step of adhering a void-free, thermoplastic skin layer to a core layer surface, the void-free skin layer being of a thickness such that the asperities of the core layer are not, at least substantially, manifest.