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- (72) Inventors ROBERT CLIVE LANSBURY and THOMAS GEOFFREY HEGGS

(54) FILM-FORMING OLEFIN POLYMER COMPOSITIONS

(71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, Imperial Chemical House, Millbank, London, SW1P 3JF, a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a thermoplastic, film-forming composition, to a self-supporting film formed from the composition, and in particular to a multiple-layer film at least one layer of which is formed from said composition.

Accordingly, the invention provides a thermoplastic, film-forming composition comprising a blend of

- (a) a propylene-ethylene copolymer, the propylene content of the copolymer being from 95 to 99.75% by weight of the copolymer, and
- (b) an alpha-olefin polymer comprising a homopolymer of an alpha-olefin containing from 4 to 10 carbon atoms in its molecule or a copolymer thereof with up to 10% by weight of the alpha-olefin copolymer of an alpha-olefin containing from 2 to 10 carbon atoms in its molecule,

the alpha-olefin polymer constituting from 41 to 90% by weight of the composition.

The invention also provides a self-supporting film formed from a thermoplastic composition comprising a blend of

- (a) a propylene-ethylene copolymer, the propylene content of the copolymer being from 95 to 99.75% by weight of the copolymer, and
- (b) an alpha-olefin polymer comprising a homopolymer of an alpha-olefin containing from 4 to 10 carbon atoms in its molecule or a copolymer thereof

with up to 10% by weight of the alpha-olefin copolymer of an alpha-olefin containing from 2 to 10 carbon atoms in its molecule,

the alpha-olefin polymer constituting from 41 to 90% by weight of the composition.

By a "self-supporting" film is meant a film capable of independent existence in the absence of a supporting substrate.

The invention further provides a multiple-layer film comprising a substrate layer of a homo- or co-polymer of an alpha-olefin the molecule of which contains from 2 to 6 carbon atoms, and, on at least one surface of the substrate, a layer formed from a composition comprising a blend of

- (a) a propylene-ethylene copolymer, the propylene content of the copolymer being from 95 to 99.75% by weight of the copolymer, and
- (b) an alpha-olefin polymer comprising a homopolymer of an alpha-olefin containing from 4 to 10 carbon atoms in its molecule or a copolymer thereof with up to 10% by weight of the alpha-olefin copolymer of an alpha-olefin containing from 2 to 10 carbon atoms in its molecule,

the alpha-olefin polymer constituting from 41 to 90% by weight of the composition.

The invention still further provides a method of producing a multiple-layer film comprising forming a substrate of the hereinbefore defined alpha-olefin polymer, and applying to at least one surface of the substrate a composition, as hereinbefore defined.

Suitable substrate materials are homo- and copolymers of alpha-olefins—such as ethylene, propylene, butene-1, 4-methylpentene-1, and hexene-1. High density polyethylene, i.e. polyethylene of density in excess of 0.949 g/cm³ and preferably less than 0.99 g/cm³, is a suitable substrate material, although for pack-

- 8 JAN 1981

- aging applications a preferred substrate is a propylene homo- or block co-polymer, the latter containing, for example, up to 15% by weight of the copolymer of another copolymerisable alpha-olefin containing 2 or from 4 to 6 carbon atoms in its molecule. The content of the other alpha-olefin is suitably from 0.25 to 15%, preferably from 0.5 to 10%, by weight of the copolymer.
- The C_4 to C_{10} alpha-olefins from which the second polymer component [component (b)] of our compositions is suitably formed include pentene-1, hexene-1, heptene-1, and decene-1, although butene-1 is preferred. When the second component is a copolymer, the content of the C_2 to C_{10} alpha-olefin is suitably from 0.25 to 10%, preferably from 0.5 to 6% by weight of the copolymer.
- The content of ethylene in the propylene-ethylene copolymer [component (a)] should not be less than 0.25% by weight of the weight of the copolymer, because below that content the softening temperature of the copolymer is so high as to produce a film with a very high threshold sealing temperature, i.e. the lowest heat-sealing temperature at which seals of acceptable strength are obtained. Likewise, to prevent undue disruption of the crystalline structure of the propylene-ethylene copolymer, the content of ethylene should be maintained at a level not exceeding 5% by weight of the copolymer. In practice, the content of ethylene is suitably from 0.5 to 4.5%, preferably from 2 to 4%, by weight of the copolymer.
- To ensure the production of heat-sealable films which exhibit acceptable heat-seal strengths, the alpha-olefin polymer content [component (b)] should be within a range of from 41 to 90% by weight of the composition, preferably within a range of from 42 to 80% by weight of the composition. However, to ensure the production of films exhibiting acceptable optical characteristics, such as wide angle haze and gloss, and particularly when the second polymer component is a butene-1 homo- or co-polymer, we prefer that the content of the second polymer should be within a range of from 45 to 65% by weight of the composition. Desirable heat-sealing characteristics are achieved when the butene-1 homo- or co-polymer content is 50% by weight of the composition.
- Suitably, the propylene-ethylene copolymer, in granular form prior to incorporation into the film-forming composition, has a Melt Flow Index, measured in accordance with ASTM/D1238-65T (condition N), of between 10 and 400 g/10 minutes, preferably between 10 and 200 g/10 minutes, and particularly preferably between 60 and 80 g/10 minutes. Suitable propylene-ethylene copolymers, therefore, have a molecular weight (weight average) of from about 350,000 to about 120,000, and preferably from about 350,000 to about 140,000.
- Suitably, the second polymer, component (b), in granular form prior to incorporation into the film-forming composition, has a Melt Flow Index, measured by the hereinbefore specified technique, of less than 50 g/10 minutes, and preferably, to ensure acceptable heat-sealing characteristics in films formed from the composition, between 5 and 40 g/10 minutes, particularly preferably between 10 and 25 g/10 minutes.
- Although the propylene-ethylene copolymer and the second copolymer component employed in the film-forming compositions may be described as random copolymers, we believe that the copolymerisation may not be random in an exact statistical sense. Thus, although in the production of our copolymers the respective comonomers are usually fed simultaneously to a polymerisation reactor for at least a substantial part of the reaction time, it is probably easier for either the propylene or other alpha-olefin monomer units to link up to the growing polymer chain immediately adjacent to an identical monomer unit, so that there may be a tendency for short sequences of identical monomer units to form preferentially in the polymer chain.
- Formation of our copolymers is conveniently effected in the presence of a "stereospecific" catalyst by which is meant a catalyst which will under equivalent conditions polymerise propylene in the absence of other monomers, to polypropylene which is at least 70% insoluble in boiling heptane. Catalysts capable of doing this comprise a transition metal compound and an organometallic activator; they are well known in the art—see for example, "Linear and Stereoregular Addition Polymers" by Gaylord and Mark, published by Interscience in 1959. Particularly useful compounds of this type are trivalent titanium compounds activated with aluminium alkyl compounds. We prefer to use titanium trichloride obtained by reduction of titanium tetrachloride either with an aluminium alkyl compound or with aluminium metal, with an aluminium alkyl halide as activator. A particularly useful and highly stereospecific catalyst is obtained by reducing titanium tetrachloride in an inert hydrocarbon diluent at a temperature of from -20°C . to $+20^{\circ}\text{C}$. with an aluminium alkyl sesquichloride, preferably by adding the sesquichloride gradually to the titanium tetrachloride dissolved in the hydrocarbon, the titanium trichloride material so obtained being recovered, optionally washed or heated one or more times between $40-150^{\circ}\text{C}$., or both, and used as a catalyst in the presence of an aluminium dialkyl chloride, such as diethylaluminium chloride, as activator.
- Catalyst components may be introduced

into the polymerisation zone as solutions or suspensions in hydrocarbon diluent. Polymerisation is normally carried out at temperatures of from 20 to 75° C. and may be effected

5 in the presence of a diluent such as an inert hydrocarbon, propylene itself, or a mixture of propylene with an inert hydrocarbon.

A range of polymerisation pressures may be used, but it is necessary to control the ratio of the partial pressures of propylene and other alpha-olefin in the reaction vessel so as to produce copolymer of the required composition. Polymerisation is carried out in the absence of substantial quantities of air and water as these materials inhibit the catalysts used, but a chain transfer agent, such as hydrogen, may be present to control the molecular weight of the copolymer. When polymerisation is complete, the reaction may be stopped, for example, by adding an alcohol, such as isopropanol, and the catalyst residues removed by washing with a suitable reagent, e.g. water or more isopropanol, or mixtures of acetylacetone and lower alcohols.

25 The production of these copolymers may be effected by the technique similar to that disclosed in our British Patent No. 1,084,953 for the production of propylene copolymers rich in butene-1, which disclosure is incorporated herein by reference.

30 Propylene-ethylene copolymers for use in the film-forming compositions of the present invention are substantially crystalline, and suitably exhibit a degree of crystallinity, as hereinafter defined, of from 50 to 65%, preferably from 57.5 to 60%.

The degree of copolymer crystallinity referred to herein is determined by a technique basically similar to that described by Natta, Corradini, and Cesari, *Rend. Accad. Naz. Lincei* 1957, 22, 11. Thus, an X-ray diffraction scan of a copolymer sample, thermally cycled as hereinafter described, is obtained using a Philips PW 1010 diffractometer, 45 working in transmission, with Cu-K α radiation and pulse height discrimination. A straight background base line is drawn in on the resultant "crystalline" diffraction spectrum between θ of 4° and 16°, where θ is the Bragg angle, and the diffraction spectrum of a 100% amorphous polypropylene sample is congruently superimposed on the "crystalline" spectrum in this region of θ 50 so that the amorphous spectrum grazes the minimum of the "crystalline" spectrum occurring at θ of approximately 7.75°, a minimum at this point being indicative of the absence of crystalline modifications other than the alpha or monoclinic crystalline modifications of polypropylene. The total area 60 between the "crystalline" spectrum and the amorphous spectrum in the specified range of θ is equated to the intensity of crystalline scattering (C), and the area between the amorphous spectrum and the base line is

equated to the intensity of amorphous scattering (A). The percentage degree of crystallinity is then defined as $100C/(C+A)$, with no corrections.

The thermal history of the copolymer samples prior to determination of crystallinity is of importance, and to achieve consistent results within the aforementioned definition samples are prepared by pressing, at 210° C., thin plates from the copolymer powder as originally made, and cooling the plate at a moderate rate. A specimen (10 mg) is cut from the plate, and placed in a standard aluminium pan (6.35 mm diameter) the latter then being placed in a cavity in a brass block sealed by a thick brass lid, the sealed block together with the lid measuring approximately 15 x 100 x 120 mm, and heated by immersion of the sealed block in an oil bath of approximately 5 litres capacity maintained at a temperature of 160° C. 85

Introduction of the brass block reduces the temperature of the bath, and slow heating of the bath is therefore continued until a temperature of 160° C. is recorded on a thermocouple positioned within the brass block. This final heating process occupies some 15 minutes, the desired temperature of 160° C. being reached asymptotically and the increase from 150° C. to 160° C. occupying a period of about 10 minutes. Cooling is then started immediately, and continued at a controlled rate of 6° C. per hour until the sample attains ambient temperature. The cooled sample is then finely chopped, and placed in a thin-walled PANTAK glass tube of 2 mm diameter for X-ray examination. 100

As hereinbefore described, the propylene-ethylene copolymers employed in the compositions of the present invention when examined by the aforementioned technique, before the copolymers have been incorporated into the composition, exhibit a degree of crystallinity of from 50 to 65%. By way of comparison, a typical commercially available propylene homopolymer exhibits a degree of crystallinity, determined by the same technique, of the order of 70%. 110

Commercially available propylene homopolymer normally crystallises in the alpha or monoclinic form when slowly cooled, e.g. at a rate of about 6° C. per hour, from its melting temperature in the absence of added nucleating agents. Under certain extreme conditions, for example under the application of exceedingly high pressures, it has been observed that a proportion of the gamma or triclinic crystalline modification becomes associated with the normal alpha or monoclinic modification, but that on orienting the product, for example by cold drawing or by drawing from the melt, the gamma modification reverts to the alpha-form so that the crystalline content of the oriented polypropylene consists substantially entirely of 130

the alpha or monoclinic crystalline modification. It is propylene homopolymers of this kind, having a crystalline content substantially entirely of the alpha or monoclinic modification, which are preferred for use in the substrate layer of the multiple-layer films of the present invention.

We have also observed that random copolymers of propylene with small proportions, e.g. up to about 15% by weight of the copolymer, of ethylene, butene-1, and higher mono-olefins, develop a relatively high proportion of the gamma crystalline form mixed with the alpha-form on slow cooling from a melt. For example, a series of propylene-ethylene copolymers, in which the content of ethylene by weight of the copolymer ranged from 2 to 4.5%, exhibited a degree of crystallinity, as hereinbefore defined, of from about 60 to 57.5%, the crystalline content of the copolymers consisting essentially of a mixture of the alpha and gamma modifications, the latter constituting from about 22 to 46% of the total crystalline content. No trace of the beta or hexagonal crystal modification was observed.

X-ray diffraction examination of oriented multiple-layer films formed in accordance with the present invention, and containing one or more layers of a copolymer of the kind referred to in the immediately preceding paragraph, revealed no trace of the gamma crystalline modification, indicating that the latter had reverted to the alpha modification which constituted the entire crystalline content of the examined copolymer layers. Preferably, therefore, the crystalline content of the substrate layer, and of the, or each, surface layer of multiple-layer films according to the present invention is of the same crystalline form.

Formation of the compositions of the present invention is conveniently achieved by dry blending, for example in a tumbler blender, the appropriate quantities of the propylene-ethylene copolymer and the alpha-olefin polymer component (b). However, we prefer to employ a melt blending technique to ensure the production of a homogeneous blend so that films formed therefrom may exhibit improved optical characteristics.

Self-supporting films are suitably formed from the compositions of the present invention in any conventional manner—for example, by extrusion, pressing, or solvent- or melt-casting techniques. However, the compositions are particularly suitable for incorporation into multiple-layer films, the composition constituting at least one layer adhered to a substrate layer.

Application of the heat-sealable composition layer onto the substrate is conveniently effected by any of the laminating or coating techniques conventionally employed in the production of composite films. Thus pre-

formed sheets of the substrate material and of the composition may be laminated together, preferably with the use of adhesives—for example, by passing the components of the laminate together between pressure rollers maintained at suitable temperatures. Alternatively, the composition may be applied to the substrate as a solution or dispersion in a suitable, preferably hot, liquid medium, such as toluene, *o*, *m* or *p*-xylene, monochlorobenzene, cyclohexane, or trichloroethylene, which may subsequently be recovered and recycled for further use. Preferably, however, the composition is applied to the substrate by means of a melt- or extrusion-coating technique in which either a molten layer of the composition is extruded directly onto the surface of a preformed substrate, or the polymeric components of the substrate and composition layer are coextruded under conditions such that the component layers come into contact with each other while each is still molten.

When the composition is applied to a preformed substrate, the adhesion of the applied layer to the substrate may be improved by orienting the substrate after application of the composition.

Adhesion of the heat-sealable composition layer to the substrate may also be improved, if desired, by the use of an intermediate anchor coating applied to the substrate prior to application of the composition layer. Suitable anchor coating materials include ethylene-vinyl acetate copolymers, or ethylene-alkyl (meth)acrylate copolymers.

However, as hereinbefore described, we prefer to employ a coextrusion technique, and when two or more molten polymeric layers are coextruded and contacted with each other while still in the molten state, the need to employ intermediate anchor coatings is obviated. Suitably, the coextrusion is effected from a multi-channel die so designed that the molten polymeric components constituting individual layers of the composite film merge at their boundaries within the die to form a single composite web which is then extruded from a common die orifice. It will therefore be understood that any reference in this specification to the composition layer being "applied to", "provided on", or otherwise deposited on the substrate layer includes the formation of a multiple-layer film structure by the simultaneous coextrusion of the substrate layer and a composition layer on one or each surface of the substrate layer.

Preferably, films made in accordance with this invention are stretched to orient them. Orientation may be effected uniaxially, by stretching the film in one direction, or biaxially, by stretching the film in each of two mutually perpendicular directions in the plane of the film, and the biaxial orientation

5		70	5
10		75	10
15		80	15
20		85	20
25		90	25
30		95	30
35		100	35
40		105	40
45		110	45
50		115	50
55		120	55
60		125	60
65		130	65

be balanced or unbalanced, for example with the higher degree of orientation of an unbalanced film in a preferred direction—usually the longitudinal or machine direction. Conveniently, the polyolefin substrate material and the heat-sealable composition are co-extruded in the form of a composite tube which is subsequently quenched, reheated, and then expanded by internal gas pressure to induce transverse orientation, and withdrawn at a rate greater than that at which it is extruded to stretch and orient it in the longitudinal direction. Alternatively, a flat composition-coated film may be oriented by simultaneous or sequential stretching in each of two mutually perpendicular directions by means of a stenter, or by a combination of draw rolls and a stenter. When a composite, heat-sealable film is formed by bonding together two preformed films, only the substrate film need be oriented prior to the bonding operation.

The degree to which the film is stretched depends to some extent on the ultimate use for which the film is intended, but for a polypropylene-based packaging film we find that satisfactory tensile and other properties are developed when the film is stretched to between three and ten, preferably, seven, times its original dimensions in each of the transverse and longitudinal directions.

After stretching, the polymeric film is normally "heat-set", while restrained against shrinkage, at a temperature above the glass transition temperature of the polymer and below its melting point. The optimum heat-setting temperature can readily be established by simple experimentation, and for a polypropylene substrate, "heat-setting" is conveniently effected at temperatures in the range of from 115° to 160° C. Heat-setting may be effected by conventional techniques—for example by means of a stenter system, or by a system of one or more heated rollers as disclosed, for example, in British Patent 1,124,886.

Composite multiple-layer films in accordance with the present invention may be prepared in a range of thicknesses governed primarily by the ultimate application for which a particular film is to be employed. Films, having a total thickness in a range extending from 2.5 to 100 microns are of general utility, but for packaging applications we prefer to employ film of from about 10 to 50 microns in total thickness.

The ratio of substrate to composition layer thickness may vary within a wide range, although we prefer that the thickness of the composition layer should not be less than 0.2% (preferably 0.5%), nor greater than 50% of that of the substrate.

Hitherto, in multiple-layer films having an external layer comprising a propylene/butene-1 copolymer layer it has been observed that

a desirably high hot seal strength is achieved only when the thickness of the propylene copolymer layer is maintained within a narrow range extending from about 0.25 to 0.45 micron. However, use of the compositions of the present invention comprising a blend of a propylene-ethylene copolymer and an alpha-olefin polymer component enables acceptably high hot seal strengths to be achieved when the thickness of the composition layer is as low as about 0.1 micron.

A further advantage of replacing the propylene/butene-1 copolymer surface layer with a layer of a composition according to the present invention is that the thickness of the surface layer required to yield acceptable "cold" heat-seal strengths is reduced from at least about 0.2 micron to 0.1 micron, or less. Additionally, the threshold sealing temperature can be reduced by several degrees Centigrade.

If desired, a multiple-layer film may be formed by providing both surfaces of the substrate with a heat-sealable layer of a composition according to the present invention, and the thicknesses of the two composition layers may be the same or different. For example a substrate film of 25 microns thickness is conveniently provided with a composition layer of 0.75 micron thickness on one surface and with a composition layer of 0.25 micron thickness, or less, on the other surface. Alternatively, one surface of the substrate may be uncoated, or may be coated with a layer of a different composition depending on the properties required of the film for a particular application.

If desired, for example to improve print receptivity, the non-seal-forming surface of a multiple-layer film having a heat-sealable layer of a composition according to the present invention may be subjected to a chemical treatment, or to a physical treatment which oxidises the surface of the film. For example, the surface may be treated with chromic acid in sulphuric acid, with hot nitric acid, or with ozone. Alternatively, the film surface may be exposed to ionising radiation, or to a thermal flame for a sufficient time to effect superficial oxidation without distortion of the surface. A preferred treatment, because of its simplicity and effectiveness, is to subject the film surface to a high voltage electric stress accompanied by corona discharge, but we have observed that such treatment tends to adversely affect the attainable hot and cold seal strengths.

If desired, the "cold" heat-seal strength and/or the hot seal strength of multiple-layer films having a surface layer of a composition according to the present invention may be improved by incorporating into the substrate layer a portion of scrap polymeric material, both substrate and composition layer(s), reclaimed from a previously pro-

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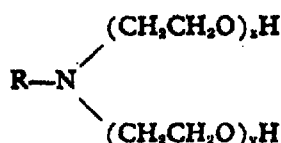
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duced multiple-layer film. Conveniently, the reclaimed material constitutes 25 to 50%, preferably from 30 to 45%, by weight of the substrate layer, but reclaim levels within an extensive range on either side of this value can be tolerated. Significantly improved seal strengths have, in fact, been observed when the substrate layer is formed entirely from reclaimed material.

The compositions of the present invention and one or more of the layers of the composite films formed therefrom may conveniently contain any of the additives conventionally employed in the manufacture of thermoplastic films. Thus, additives such as dyes, pigments, lubricants, anti-static agents, antioxidants, anti-blocking agents, surface-active agents, slip aids, stiffening aids, gloss-improvers, prodegradants, and ultra-violet light stabilisers may be employed. Films are conveniently rendered anti-static by the incorporation therein of from 0.1 to 1.0% by weight of the film of one or more substances of formula



wherein $x + y$ is from 2 to 5 and R is a monovalent aliphatic radical containing from 12 to 22 carbon atoms, and from 0.01 to 0.5% based on the weight of the film of a fatty acid amide derived from a fatty acid containing from 10 to 22 carbon atoms, as disclosed in our British Patent 1,034,337.

Composite films formed with a surface layer of a composition in accordance with the present invention are heat-sealable at comparatively low temperatures, for example at temperatures of the order of 120° C. and lower, to provide a strong, hermetic seal. In addition, the heat-sealable composition layer adheres securely to the substrate material, exhibits desirable scratch- and scuff-resistance, and is resistant to sticking to the jaws of the heat-sealing equipment.

The elimination of jaw-sticking is of considerable importance to the packaging industry where the intermittent sticking of a packaging film to the jaws of heat-sealing equipment employed to form individual packages by sealing the film to itself, can impede or halt the uniform passage of film through the equipment, thereby disrupting the packaging operation. The problem is of particular concern in relation to packaging machines of the so-called vertical "form and fill" type.

Films formed from the compositions of the present invention are suitable for use in the packaging industry, particularly for the formation of flexible packages for foodstuffs and the like. They may, however, be employed

in any other application for which a heat-sealable polyolefin film is required.

The hot seal strength rating of a film formed from a composition of the present invention is determined by means of a calibrated strip of spring steel of approximately 25 mm width and 400 mm length. Calibration of the spring steel strip is effected by folding the strip over upon itself about its mid-point, somewhat in the form of a "C" with elongated limbs, and gradually applying an increasing load in the form of weights positioned on the uppermost limb adjacent the free end thereof until the free ends of the spring just make contact with each other. The required loading of the selected spring was 100 grammes, and the spring is therefore rated as a "100 gramme spring".

A strip of a multiple-layer film having a width of 25 mm and a length of about 150 mm, is folded over upon itself for testing so that portions of the surface layer formed from a composition of the present invention are in contact with each other, and the free ends of the film strip are secured by a suitable adhesive to the outermost surface of the free ends of the previously calibrated spring strip. The latter is maintained under manual compression so that the free ends of the spring strip remain in contact until the opposed surfaces of the film strip are sealed together between the heated jaws of a Sentinel Heat Sealer (Model No. 12AS)—(the word Sentinel is a registered Trade Mark), the free ends of the spring strip being positioned outside but immediately adjacent the edges of the sealing jaws. The depth "y" of the jaws, aligned parallel to the longitudinal axis of the film strip, is 25 mm, so that a square heat-seal of side 25 mm is formed between the opposed surfaces of the film strip.

Upon closure of the sealing jaws the manual compression force is removed from the steel strip so that immediately upon retraction of the sealing jaws to the open position after formation of the heat-seal, the free ends of the steel strip are allowed to spring apart thereby tending to rupture the newly created heat-seal. The depth "x" of the opening thereby induced in the seal, parallel to the length of the film strip, provides a measure of the strength of the seal, the degree of seal opening being expressed in the form $x \times 100/y\%$. The test procedure is then repeated on other samples of film.

In contrast to the hot seal strength which is determined before the thermal energy employed to form the heat-seal has dissipated, the "cold" heat-seal strength of the film is determined after the seal has cooled to ambient temperature, and the full potential strength of the seal has developed.

To determine the "cold" heat-seal

strength, two strips of a multiple-layer film having at least one surface layer formed from a composition of the present invention were placed with the composition surfaces in contact with each other, and the ends of the strips sealed together between the jaws of a Sentinel Heat Sealer, Model No. 12AS, only one jaw of the unit being heated. The strength of the seal was measured by cutting from the sealed-together strips of film, which had been allowed to cool to ambient temperature, a strip 25 mm wide and some 100 mm long, the heat-seal extending across the entire width of the strip and having an area of some 25 x 25 mm. The unsealed ends of the strip were then attached respectively to a dynamometer and a gripping surface, a load was applied to the gripping surface, and the minimum load required to separate the strips in the sealed area was recorded by the dynamometer, and expressed as a seal strength of x g/25 mm, where x was the load in grammes.

The invention is illustrated by reference to the drawings accompanying the provisional specification in which:

Figure 1 depicts a side elevation of a portion of a propylene homopolymer base film 1 having a thin layer 2, formed from a blend of a propylene-ethylene copolymer and a butene-1 polymer, on one surface thereof, and

Figure 2 depicts a side elevation of a similar substrate film 1 having a thin layer of the blend 2, 3 on each surface thereof.

The invention is further illustrated by reference to the following Examples.

EXAMPLES 1 to 6.

These are comparative Examples.

From a triple channel annular die were co-extruded a propylene homopolymer, and a propylene/butene-1 random copolymer containing 12% by weight of butene-1 and having a granular Melt Flow Index (MFI), as hereinbefore defined, of approximately 65 g/10 minutes, so as to form a polypropylene tube the internal and external surfaces of

which were coated with a layer of the propylene/butene-1 copolymer. The propylene homopolymer contained, as anti-oxidant, 0.1% by weight of 1,3,5 - tri(3,5 - ditertiarybutyl - 4-hydroxybenzyl)2,4,6 - trimethylbenzene.

The coated polypropylene tube was cooled by passage over a mandrel within the tube, and externally quenched by passage through a water bath surrounding the mandrel, heated to stretching temperature, expanded by internal gas pressure, and withdrawn from the expansion zone at a rate greater than that at which it was fed thereto, so that the tube was stretched to seven times its original dimensions in both the direction of extrusion and in a direction transverse thereto.

The stretched tubular film was then opened out to form flat film which was subsequently heat-set at a temperature of 125° C. on a matt-surfaced roller heat-setter of the kind described in British Patent 1,124,886. The thickness of the copolymer layer on each surface of the resultant film was 0.13 micron, the substrate having a thickness of about 25 microns.

This procedure was repeated several times with appropriate adjustment of the rate of copolymer flow to produce a series of films in which the thickness of the copolymer layer on each surface of the substrate was respectively 0.22, 0.31, 0.39, 0.50, and 0.59 micron.

A sample of each film was heat-sealed on a Sentinel Heat Sealer, Model No. 12AS, operating at a jaw temperature of 130° C., jaw pressure of 15 psi, and jaw closure time of 2 seconds. The hot seal strength of the resultant seals was determined, as hereinbefore described, using a steel strip spring rated at 100 grammes.

The "cold" heat-seal strength (at ambient temperature) of the films was assessed by the dynamometer technique, as hereinbefore described, on a sample of each film sealed at 120° C. on a Sentinel Heat Sealer, Model No. 12AS, operating at a jaw pressure of 15 psi and jaw closure time of 2 seconds.

The results are recorded in the accompanying Table.

TABLE

Example No.	Surface layer thickness (μm)	Hot seal strength % opening (a)	Cold seal strength (g/25 mm) (b)
1	0.13	86	0
2	0.22	2	390
3	0.31	12	375
4	0.39	35	480
5	0.50	35	480
6	0.59	32	525

(a) seal formed at 130°C

(b) seal formed at 120°C

EXAMPLES 7 to 12.

These are also comparative Examples.

- 5 The procedure of Examples 1 to 6 was repeated except that in each case the propylene/butene-1 copolymer was replaced by a propylene-ethylene random copolymer

containing 3% by weight of ethylene and having an MFI of approximately 35 g/10 minutes.

Results of seal strength measurements are recorded in the accompanying Table.

TABLE

Example No.	Surface layer thickness (μm)	Hot seal strength % opening (a)	Cold seal strength (g/25 mm) (b)
7	0.13	100	0
8	0.22	100	0
9	0.31	100	0
10	0.39	100	0
11	0.50	100	0
12	0.59	100	0

(a) seal formed at 130°C

(b) seal formed at 120°C

- 15 The above results show that the propylene-ethylene copolymer is inferior to the propylene/butene-1 copolymer in terms of both the "hot" and "cold" seal strengths.

EXAMPLES 13 to 18.

- 20 The procedure of Examples 1 to 6 was repeated except that in each case the propyl-

ene/butene-1 copolymer was replaced by a blended composition comprising 50 parts by weight of the propylene-ethylene random copolymer of Examples 7 to 12 and 50 parts by weight of polybutene-1.

Results of seal strength measurements are recorded in the accompanying Table.

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TABLE

Example No.	Surface layer thickness (μ m)	Hot seal strength % opening (a)	Cold seal strength (g/25 mm) (b)
13	0.13	57	130
14	0.22	40	530
15	0.31	100	530
16	0.39	100	490
17	0.50	100	490
18	0.59	100	470

(a) seal formed at 130°C

(b) seal formed at 120°C

These results demonstrate that blended compositions according to the present invention enable an acceptable "hot" seal strength under the specified conditions to be achieved with a surface layer thickness of the order of 0.13 micron (57% opening) in contrast to the 86% and 100% opening exhibited respectively by the propylene/butene-1 copolymer layer and the propylene-ethylene copolymer layer of the same thickness.

Similarly, the "cold" seal strength of the blended composition layer is superior to that of the propylene/butene-1 and of the propylene-ethylene copolymer layer.

The films also exhibit excellent optical characteristics, being clear and glossy.

WHAT WE CLAIM IS:—

1. A thermoplastic, film-forming composition comprising a blend of

- (a) a propylene-ethylene copolymer, the propylene content of the copolymer being from 95 to 99.75% by weight of the copolymer, and
- (b) an alpha-olefin polymer comprising a homopolymer of an alpha-olefin containing from 4 to 10 carbon atoms in its molecule or a copolymer thereof with up to 10% by weight of the alpha-olefin copolymer of an alpha-olefin containing from 2 to 10 carbon atoms in its molecule.

the alpha-olefin polymer constituting from 41 to 90% by weight of the composition.

2. A composition according to claim 1 wherein the propylene copolymer in granular form has a Melt Flow Index, as hereinbefore defined, of from 10 to 400 g/10 minutes.

3. A composition according to either of claims 1 and 2 wherein the propylene copolymer exhibits a degree of crystallinity, as hereinbefore defined, of from 50 to 65%. 40

4. A composition according to any one of the preceding claims wherein the crystalline content of the propylene copolymer is of monoclinic crystalline form. 45

5. A composition according to any one of the preceding claims wherein the ethylene content of the propylene copolymer is from 2 to 4% by weight of the copolymer.

6. A composition according to any one of the preceding claims wherein the alpha-olefin polymer, component (b), has a Melt Flow Index, as hereinbefore defined, of from 5 to 40 g/10 minutes. 50

7. A composition according to any one of the preceding claims wherein the alpha-olefin polymer comprises from 45 to 65% by weight of the composition. 55

8. A composition according to any one of the preceding claims wherein the alpha-olefin polymer is a polybutene-1 homopolymer.

9. A self-supporting film formed from a composition according to any one of the preceding claims.

10. A multiple-layer film comprising a substrate layer of a homo- or co-polymer of an alpha-olefin the molecule of which contains from 2 to 6 carbon atoms, and, on at least one surface of the substrate, a layer formed from a composition according to any one of claims 1 to 8. 70

11. A multiple-layer film according to claim 10 wherein the substrate layer comprises a homopolymer of propylene, or a block copolymer thereof with up to 15% by weight of the copolymer of an alpha-olefin contain- 75



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TABLE

Example No.	Surface layer thickness (μm)	Hot seal strength % opening (a)	Cold seal strength (g/25 mm) (b)
13	0.13	57	130
14	0.22	40	530
15	0.31	100	530
16	0.39	100	490
17	0.50	100	490
18	0.59	100	470

(a) seal formed at 130°C

(b) seal formed at 120°C

These results demonstrate that blended compositions according to the present invention enable an acceptable "hot" seal strength under the specified conditions to be achieved with a surface layer thickness of the order of 0.13 micron (57% opening) in contrast to the 86% and 100% opening exhibited respectively by the propylene/butene-1 copolymer layer and the propylene-ethylene copolymer layer of the same thickness.

Similarly, the "cold" seal strength of the blended composition layer is superior to that of the propylene/butene-1 and of the propylene-ethylene copolymer layer.

The films also exhibit excellent optical characteristics, being clear and glossy.

WHAT WE CLAIM IS:—

1. A thermoplastic, film-forming composition comprising a blend of

- (a) a propylene-ethylene copolymer, the propylene content of the copolymer being from 95 to 99.75% by weight of the copolymer, and
- (b) an alpha-olefin polymer comprising a homopolymer of an alpha-olefin containing from 4 to 10 carbon atoms in its molecule or a copolymer thereof with up to 10% by weight of the alpha-olefin copolymer of an alpha-olefin containing from 2 to 10 carbon atoms in its molecule.

the alpha-olefin polymer constituting from 41 to 90% by weight of the composition.

2. A composition according to claim 1 wherein the propylene copolymer in granular form has a Melt Flow Index, as hereinbefore defined, of from 10 to 400 g/10 minutes.

3. A composition according to either of claims 1 and 2 wherein the propylene copolymer exhibits a degree of crystallinity, as hereinbefore defined, of from 50 to 65%.

4. A composition according to any one of the preceding claims wherein the crystalline content of the propylene copolymer is of monoclinic crystalline form.

5. A composition according to any one of the preceding claims wherein the ethylene content of the propylene copolymer is from 2 to 4% by weight of the copolymer.

6. A composition according to any one of the preceding claims wherein the alpha-olefin polymer, component (b), has a Melt Flow Index, as hereinbefore defined, of from 5 to 40 g/10 minutes.

7. A composition according to any one of the preceding claims wherein the alpha-olefin polymer comprises from 45 to 65% by weight of the composition.

8. A composition according to any one of the preceding claims wherein the alpha-olefin polymer is a polybutene-1 homopolymer.

9. A self-supporting film formed from a composition according to any one of the preceding claims.

10. A multiple-layer film comprising a substrate layer of a homo- or co-polymer of an alpha-olefin the molecule of which contains from 2 to 6 carbon atoms, and, on at least one surface of the substrate, a layer formed from a composition according to any one of claims 1 to 8.

11. A multiple-layer film according to claim 10 wherein the substrate layer comprises a homopolymer of propylene, or a block copolymer thereof with up to 15% by weight of the copolymer of an alpha-olefin contain-

ing 2, 4, 5 or 6 carbon atoms in its molecule.

12. A multiple-layer film according to either of claims 10 and 11 wherein the crystalline content of the substrate layer, and of the, or each, surface layer is of the same crystalline form.

13. A multiple-layer film according to claim 12 wherein the crystalline content of the substrate layer, and of the, or each, surface layer is of monoclinic crystalline form.

14. A multiple-layer film according to any one of claims 10 to 13 wherein the thickness of the, or each, composition layer is from 0.2 to 50% of that of the substrate.

15. A multiple layer film according to any one of claims 10 to 14 wherein the substrate layer contains a portion of scrap polymeric material reclaimed from a previously produced multiple-layer film according to claim 10.

16. A biaxially oriented multiple-layer film comprising a substrate layer of a propylene homopolymer or a copolymer of propylene with from 0.5 to 10% of ethylene by weight of the copolymer, and, on at least one surface of the substrate, a coextruded layer formed from a blend comprising from 55 to 35% by weight of a propylene-ethylene random copolymer containing from 0.5 to 4.5%

of ethylene by weight of said random copolymer, and from 45 to 65% by weight of a butene-1 homopolymer.

17. A multiple-layer film substantially as herein described with reference to any one of Examples 13 to 18.

18. A method of producing a multiple-layer film comprising forming a substrate layer of a polymer or copolymer of an alpha-olefin, the molecule of which contains from 2 to 6 carbon atoms, and applying to at least one surface of the substrate a layer of a composition according to any one of claims 1 to 8.

19. A method according to claim 18 comprising coextruding the substrate layer and the composition layer or layers.

20. A method according to either of claims 18 and 19 comprising orienting the substrate.

21. A method according to claim 20 comprising heat-setting the oriented film.

22. A method according to claim 18 substantially as herein described with reference to any one of Examples 13 to 18.

23. A multiple-layer film made by the method of any one of claims 18 to 22.

J. L. RHIND,
Agent for the Applicants.

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