EXHIBIT U

PART 2

LINEAR ETHYLENE INTERPOLYMER BLENDS OF INTERPOLYMERS HAVING NARROW MOLECULAR WEIGHT AND COMPOSITION DISTRIBUTIONS

SPECIFICATION

Field of the Invention

The present invention relates to polymer blends of crystalline interpolymers, such as LLDPE, and more particularly to blends of such interpolymers wherein each blend component has a narrow molecular weight distribution and a narrow composition distribution selected to obtain superior properties in the blend.

Background of the Invention

Various types of polyethylene are known in the art. Low density polyethylene ("LDPE") is generally prepared at high pressure using free radical initiators and typically has a density in the range of 0.915-0.940 g/cm^3. LDPE is also known as "branched" polyethylene because of the relatively large number of long chain branches extending from the main polymer backbone.

High density polyethylene ("HDPE") usually has a density in the range of greater than 0.940 to 0.960 g/cm^3. HDPE is prepared using a coordination catalyst, e.g. Ziegler-Natta type catalysts, at low or moderate pressures, but sometimes at high pressure. HDPE is

generally linear without any substantial side chain branching. HDPE is a substantially crystalline polymer.

Linear, low density polyethylene ("LLDPE") is generally prepared in the same manner as HDPE, but incorporates a relatively minor amount of an α-olefin comonomer such as butene, hexene or octene to introduce enough short chain branches into the otherwise linear polymer to reduce the density of the resultant polymer into the range of that of LDPE. The coordination catalysts used to interpolymerize ethylene and the α-olefin generally produce an LLDPE with a relatively broad weight molecular weight distribution, i.e., Mw/Mn greater than about 3. Such LLDPE's also have relatively broad composition distribution in that the proportion of α-olefin comonomer molecules incorporated into the polymer molecules varies. Generally, the lower molecular weight polymer molecules contain a relatively higher proportion of the α-olefin comonomer than the higher molecular weight polymer molecules.

A polyethylene such as LLDPE having a broad molecular weight distribution is undesirable in many respects, depending on the desired end use application. For example, LLDPE resins known in the prior art containing relatively high molecular weight molecules are subject to orientation which results in anisotropic properties in the machine versus transverse direction of a fabrication process. On the other hand, resins containing relatively lower molecular weight molecules, in which the comonomer is invariably concentrated, tend to exhibit high block and tackiness. These lower molecular weight, highly branched molecules interfere with the proper function of certain additives compounded in the resin, increase the percentage of extractable polymer, and increase fouling in the polymerization plant. The relatively high α-olefin comonomer content of these low molecular weight polymer molecules causes such polymer molecules to be generally

amorphous and to exude to the surface of fabricated parts, thereby producing an undesirable sticky surface.

Prior art polyethylenes such as LLDPE also generally tend to have a very broad, nonuniform distribution of comonomer content, i.e. some polymer molecules have a relatively high α-olefin comonomer content while others have a relatively low content. Generally, the polymer molecules of low comonomer content are relatively more crystalline and have a high melting temperature, whereas the high comonomer content polymer molecules are more amorphous and melt at a lower temperature. The presence of a higher melting component is disadvantageous in many applications, for example where softness or clarity is desired. On the other hand, the presence of a lower melting component frequently results in a high quantity of extractables, which limit food contact applications.

Prior art blends of polyethylenes designed to improve one or more of the properties of the blend relative to its blend components or prior art polyethylene have also suffered from the drawbacks mentioned above. For example, incorporating a blend component with a high average comonomer content to reduce crystallizability generally results in an increase of extractables and adversely affects other properties so that the full advantage of the blend is not realized. Thus, there is a need to provide ethylene interpolymer blends with superior properties and in which the full advantages of blending may be realized.

Summary of the Invention

The present invention provides a blend of ethylene interpolymer components with narrow molecular weight and composition distributions selected to obtain an overall molecular weight and composition distribution in the resulting blend to impart superior properties thereto. Broadly, the blends of this invention comprise a plurality of linear ethylene interpolymer components wherein each component has a M_w/M_n value less than or equal to 3 and a composition distribution breadth index (as later

described) of 50% or higher. The blend is substantially free of blend components having both a higher average molecular weight and a lower average comonomer content than that of any other polyethylene component in the blend. The components for the blend can be selected so that the resultant blend has plural modality with respect to molecular weight distribution, comonomer content, or both.

In another aspect, the components for the blend are linear ethylene interpolymers having the narrow molecular weight and composition distributions mentioned above and the blend components are selected from one of the groups consisting of: (1) linear ethylene interpolymer blend components having substantially the same average molecular weight but different average comonomer contents; (2) linear ethylene interpolymer blend components having substantially the same average comonomer content but different average molecular weights; and (3) linear ethylene interpolymer blend components having different average molecular weights and comonomer contents in which the blend components, taken serially in order of increasing average molecular weight, have an increasing comonomer content.

In still another aspect, the linear ethylene interpolymer blend components have the narrow molecular weight and composition distribution mentioned above, and for the linear ethylene interpolymer blend components taken serially in order of increasing average molecular weight, then each succeeding component has substantially the same or a higher average comonomer content than each preceding component in said series.

In another aspect, the invention provides a linear ethylene interpolymer having plural modality with respect to comonomer content, a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and an overall composition distribution breadth index less than 50%.

In still another aspect, the invention provides a linear ethylene interpolymer having plural modality with respect to molecular weight so that the blend has broad overall molecular weight distribution such that $M_w/M_n > 3$ and a composition distribution breadth index greater than or equal to 50%.

In still another aspect, the invention provides a linear ethylene interpolymer of plural modality with respect to both comonomer content and molecular weight, comprising a plurality of components having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ for each fraction and each component taken serially in order of increasing average molecular weight, has an increasing average comonomer content.

In a still further aspect of the invention, there is provided a linear ethylene interpolymer blend of plural modality with respect to both comonomer content and molecular weight which comprises a plurality of components having a composition distribution breadth index of 50% or more, wherein the components taken serially in order of increasing comonomer content, have an increasing average molecular weight.

Brief Description of the Drawings

Fig. 1 is a schematic illustration of different blends made from poly(ethylene-co-α-olefin) blend components having narrow molecular weight and composition distributions.

Fig. 2 illustrates the broad molecular weight distribution and broad composition distribution of a typical prior art LLDPE.

Fig. 3 illustrates the narrow molecular weight distribution and narrow composition distribution of an exemplary blend component used in the present invention.

Fig. 4 illustrates the molecular weight distribution and composition distribution of an exemplary LLDPE blend

blend components have about the same molecular weight but differing comonomer contents.

Fig. 5 illustrates the molecular weight distribution and composition distribution of an exemplary LLDPE blend according to another embodiment of the invention in which the blend components have about the same comonomer content but differing molecular weights.

Fig. 6 illustrates the molecular weight distribution and composition distribution of an exemplary LLDPE blend according to yet another embodiment of the invention in which the conomoner contents of the blend components increases as the molecular weight increases.

Description of the Preferred Embodiments

The linear ethylene interpolymer blend components in the present invention are higher interpolymers of a major proportion of ethylene and a minor proportion of comonomer. The ethylene is generally polymerized in a proportion of 70-100, typically 70-97, and often 70-80, 80-90 or 90-95, mole percent of the interpolymerized monomers with 0-30, typically 3-30, and often 20-30, 10-20 or 5-10, mole percent comonomer. Contemplated blend components generally include elastomer blend components in the density range of about 0.85-0.900 g/cm^3, very low density polyethylene blend components in the density range of about 0.900-0.915 g/cm^3, and linear, low density polyethylene blend components in the density range of about 0.915-0.940 g/cm^3. Ethylene interpolymers having a density in the high density polyethylene range above about 0.940 g/cm^3 are also contemplated as being suitably employed in the invention.

Suitable comonomers interpolymerized with the ethylene to obtain the ethylene interpolymer blend components herein generally include monomers which may be copolymerized with ethylene to obtain the molecular weight distribution and comonomer distribution desired in the

α-olefins having 3 to about 12 carbon atoms, such as propylene, 1-butene, 1-pentene, 1-hexene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-octene, 1-decene, 1-dodecane and the like. Other suitable comonomers include vinyl cyclohexane, norbornene, vinyl cyclohexene, and other diene comonomers such as 1,3-butadiene, 1,4-hexadiene, 4-methyl-1,4-hexadiene, 5-methyl-1,4-hexadiene, 1,5-hexadiene and the like. The ethylene interpolymer may include one or more of such comonomers, i.e. it may be a copolymer, terpolymer, etc.

The molecular weight of the LLDPE blend components may range from one thousand to one million or more depending on the particular end use, preferably 10^4-10^6, and especially $2 \times 10^4 - 5 \times 10^5$. As used herein, the terms "average molecular weight" and "molecular weight" refer to weight average molecular weight unless otherwise indicated. The linear polyethylene blend components preferably have a narrow molecular weight distribution (MWD). By "narrow MWD" is meant that the ratio of the weight average molecular weight (M_w) to the number average molecular weight (M_n) is less than or equal to 3.0. Particularly preferred are the linear polyethylene blend components having a very narrow MWD, i.e. M_w/M_n less than or equal to 2.5, and especially less than or equal to 2.0. Molecular weight distributions of ethylene interpolymers are readily determined by techniques known in the art, such as, for example, size exclusion chromatography.

The linear polyethylene blend components preferably have a composition distribution (CD) such that the composition distribution breadth index (CDBI) is at least 50%. The CDBI is defined as the weight percent of the copolymer molecules having a comonomer content within 50 percent of the median total molar comonomer content. The CDBI of linear polyethylene, which does not contain a comonomer, is defined to be 100%. The CDBI of a copolymer is readily calculated from data obtained from techniques known in the art, such as, for example, temperature rising

elution fractionation as described, for example, in U.S. Patent Application Serial No. 151,350 or in Wild et al., J. Poly. Sci, Poly. Phys. Ed., vol. 20, p. 441 (1982). Unless otherwise indicated, terms such as "comonomer content", "average comonomer content" and the like refer to the bulk comonomer content of the indicated interpolymer blend, blend component or fraction on a molar basis.

A graphical illustration of an exemplary narrow MWD, narrow CD blend component is seen in Fig. 3. In this three-dimensional figure, the Y-axis is the molecular weight, the X-axis is the molar comonomer content, and the Z-axis represents the incidence or weight proportion of molecules. As can be seen, the MWD and the CD of the blend component are narrow and appear as relatively sharp peaks in Fig. 3. In contrast, the MWD/CD diagram for a typical conventional LLDPE seen in Fig. 2 shows a broad MWD and a broad CD, and the comonomer content tends to decrease as the molecular weight increases.

In the blends of the present invention, one or more of the properties of the blend are improved by appropriate selection of narrow MWD, narrow CD blend components. In one embodiment, for example, tear strength may be controlled by blending linear polyethylene resins having about the same average molecular weight but with different average comonomer contents. Such a blend is illustrated as line B in Fig. 1. In another embodiment, the comonomer contents of the linear polyethylene blend components are the same, but molecular weights are varied, as illustrated by line C in Fig. 1. In still further embodiments illustrated by lines D, E and F in Fig. 1, the blend components taken serially in order of increasing molecular weight, or in order of increasing molar comonomer content, have the same or higher comonomer content or molecular weight, respectively.

As used herein, two or more blend components have substantially

MWD of the blend thereof is similarly narrow to the MWD of each blend component, i.e. the value of M_w/M_n of the resulting blend is less than or equal to about 3.0, preferably less than about 2.5. Conversely, two or more blend components have a different average molecular weight if the overall M_w/M_n of the resulting blend is relatively greater than for each such blend component, i.e., the M_w/M_n of the blend is greater than 3.0. As used herein, two or more blend components have a different comonomer content if the overall CDBI of the resulting blend is relatively less than that of each such blend component, i.e., the overall CDBI of the blend is less than 50%. Conversely, two or more blend components have substantially the same molar comonomer content if the resulting CD of the blend thereof is similarly narrow with respect to each blend component, i.e., the resulting blend has a CDBI of 50% or greater. It is readily appreciated that the CD and MWD of a blend can depend on the relative proportions of each blend component employed therein. It is specifically contemplated that blend components may have the "same" molecular weight for purposes of one blend, but not for the purpose of another blend, e.g. wherein the components would result in the blend having an MWD less than or greater than 3.0 depending on the proportion of each blend component. Similarly, blend components may have a "different" comonomer content for purposes of one blend, but not for the purposes of another blend, e.g. wherein the components would result in the final blend having CDBI less than or greater than 50% depending on the proportion of each blend component.

The molecular weight and composition distribution of a bimodal blend of the invention is illustrated graphically in Fig. 4. It is seen from this MWD/CD diagram that the comonomer content of each of the blend components is different, while the molecular weight of each blend component is about the same. This blend

graph for typical conventional LLDPE is seen in Fig. 2, and line A of Fig. 1, and shows that the lower molecular weight fractions contain more of the comonomer than the higher molecular weight fractions. The lower molecular weight molecules which contain relatively high comonomer concentrations as in this conventional LLDPE can cause undesirable effects such as poor surface properties, high block and tackiness, cling development, high levels of extractables, and fouling of polymerization plants. In the present invention, such effects are minimized and properties are enhanced by providing blends of the type illustrated in Curve B, C, D, E, and F.

As an example of the embodiment of Curve B, it has been found that a 50-50 blend of a first LLDPE having a 6.4 mole% 1-butene content and a M_w of 80,400 (M_w/M_n = 2.3; CDBI = ~67%; MI 4.0 dg/min; density 0.9042 g/cm^3) with a HDPE having a 0.0 mole% 1-butene content and a M_w of 76,700 (M_w/M_n = 2.8; CDBI = ~100%; MI 5.0 dg/min; density 0.9552 g/cm^3) has an Elmendorf tear strength of 210 g/mil, synergistically enhanced in contrast to the tear strengths of 111 and 48 g/mil for the respective first and second blend components. Further, a 25-75 blend of these same components has a further enhanced Elmendorf tear strength of 227 g/mil. This result is quite surprising and unexpected because including a higher proportion of the second LLDPE resin with the lower tear strength in the blend increases the tear strength of the resulting blend, rather than decreasing the tear strength as would be expected.

In another embodiment exemplified in Fig. 5 and line C of Fig. 1, a multimodal MWD is obtained by blending linear polyethylene components having narrow molecular weight and composition distributions, and about the same comonomer content, but differing molecular weights. The molecular weight distribution of such blends improves the melt processability and rheological characteristics

stability, high shear sensitivity, reduced draw resonance. On the other hand, the optical properties of mechanical, surface and individual blend components are generally substantially retained or improved in the blend thereof, including, for example tear strength, modulus, yield strength, reduced blocking, clarity, gloss, haze and the like. Moreover, such blends have lower portions of soluble molecules than prior art copolymers having similar molecular weight distribution. Desirable molecular weight and composition distributions may be obtained by blending different components, or by polymerization of the blend components simultaneously in the same reactor or in multiple reactors.

The higher molecular weight fraction containing relatively less comonomer as in conventional LLDPE may cause an anisotropic morphology during fabrication processing known as "row nucleated" or "shish-ka-bob" morphology. This anisotropic morphology is believed to contribute to poor toughness in articles crystallized from flowing melts. In the present invention, the anisotropy may be minimized by providing a blend with lower concentrations of such higher molecular weight molecules with a relatively low comonomer content and by incorporating the comonomer in the blend components as indicated in blend B, C, D, E and F.

In another embodiment as exemplified by Fig. 5 and line D of Fig. 1, the blend includes components having narrow molecular weight and composition distributions, but differing average molecular weights and average comonomer contents. However, in contrast to conventional LLDPE as illustrated in line A of Fig. 1 and in Fig. 2, the blend of this embodiment has a greater comonomer content in the higher molecular weight fractions or blend components than in the lower molecular weight fractions or blend components. These distributions are obtained, for example, by blending narrow MWD, narrow CD linear polyethylene resins which, taken serially in order of

increasing molecular weight, have an increasing comonomer content. It is also contemplated that the blend may include two or more blend components having the same molecular weight, as illustrated by line E in Fig. 1, in which case such components would be included in the serial ordering secondarily in order of their increasing average comonomer content. Also, the presence of two or more blend components having the same comonomer content is also contemplated as being within the purview of this embodiment, as illustrated by line E in Fig. 1, provided that there is included either at least one blend component having a higher comonomer content and molecular weight or at least one blend component having a lower comonomer content and lower molecular weight than any of the blend components having the same comonomer content. In this embodiment, the blend is preferably substantially free of blend components having both a higher molecular weight and a lower comonomer content than any component present in the blend.

Such a blend has properties which are significantly different from prior art blends and conventional LLDPE resins in which the comonomer content generally decreases in proportion to increasing molecular weight components or fractions. The isotropy and toughness of films made from such blends are improved by minimizing the anisotropic shish-ka-bob or row-nucleated morphology ordinarily caused by a low concentration of comonomers present in the higher molecular weight molecules in conventional LLDPE resins. Moreover, such blends have desirable properties such as, for example, reduced blocking, reduced coefficients of friction, and lower extractables, in comparison to conventional LLDPE resins.

The linear polyethylene blend components of the invention may be prepared by use of catalyst systems of the metallocene type known to provide narrow CD/MWD resins. Cyclopentadienylide catalyst systems using a metallocene complex in conjunction with an alumoxane

cocatalyst or reaction product thereof are suitable for preparing the polymer components utilized in the blends of the invention. The metallocene catalyst may be represented by the general formula $(C_p)_mMR_nR'_p$ wherein C_p is a substituted or unsubstituted cyclopentadienyl ring; M is a Group IVB, or VB transition metal; R and R' are independently selected halogen, hydrocarbyl group, or hydrocarboxyl groups having 1-20 carbon atoms; m = 1-3, n = 0-3, p = 0-3, and the sum of m + n + p equals the oxidation state of M. Various forms of the catalyst system of the metallocene type may be used for polymerization to prepare the polymer components of the present invention including those of the homogenous or the heterogeneous, supported catalyst type wherein the catalyst and alumoxane cocatalyst are together supported or reacted together onto an inert support for polymerization by gas-phase, high pressure, slurry, or solution polymerization.

The cyclopentadienyls of the catalyst may be unsubstituted or substituted with hydrogen or hydrocarbyl radicals. The hydrocarbyl radicals may include alkyl, alkenyl, aryl, alkylaryl, arylalkyl, and like radicals containing from about 1-20 carbon atoms or where 2 carbon atoms of cyclopentadienyl are joined together to form a C_4-C_6 ring. Exemplary hydrocarbyl radicals include methyl, ethyl, propyl, butyl, amyl, isoamyl, hexyl, isobutyl, heptyl, octyl, nonyl, decyl, cetyl, 2-ethylhexyl, phenyl and the like. Exemplary halogen substituents include chlorine, bromine, flourine and iodine. Of these halogen atoms, chlorine is preferred. Exemplary hydrocarboxy radicals are methoxy, ethoxy, propoxy, butoxy, amyloxy and the like. Illustrative, but non-limiting examples of the metallocene catalyst useful in preparing the polymers of the present invention include bis(cyclopentadienyl)titanium dimethyl, bis(cyclopentadienyl)titanium diphenyl,

bis(cyclopentadienyl)zirconium dimethyl,
bis(cyclopentadienyl)zirconium diphenyl,
bis(cyclopentadienyl)hafnium dimethyl and diphenyl,
bis(cyclopentadienyl)titanium di-neopentyl,
bis(cyclopentadienyl)zirconium di-neopentyl,
bis(cyclopentadienyl)titanium dibenzyl,
bis(cyclopentadienyl)zirconium dibenzyl,
bis(cyclopentadienyl)vanadium dimethyl; the mono alkyl metallocenes such as bis(cyclopentadienyl)titanium methyl chloride, bis(cyclopentadienyl)titanium ethyl chloride, bis(cyclopentadienyl)titanium phenyl chloride, bis(cyclopentadienyl)zirconium methyl chloride, bis(cyclopentadienyl)zirconium ethyl chloride, bis(cyclopentadienyl)zirconium phenyl chloride, bis(cyclopentadienyl)titanium methyl bromide, bis(cyclopentadienyl)titanium methyl iodide, bis(cyclopentadienyl)titanium ethyl bromide, bis(cyclopentadienyl)titanium ethyl iodide, bis(cyclopentadienyl)titanium phenyl bromide, bis(cyclopentadienyl)titanium phenyl iodide, bis(cyclopentadienyl)zirconium methyl bromide, bis(cyclopentadienyl)zirconium methyl iodide, bis(cyclopentadienyl)zirconium ethyl bromide, bis(cyclopentadienyl)zirconium ethyl iodide, bis(cyclopentadienyl)zirconium phenyl bromide, bis(cyclopentadienyl)zirconium phenyl iodide; the trialkyl metallocenes such as cyclopentadienyltitanium trimethyl, cyclopentadienyl zirconium triphenyl, and cyclopentadienyl zirconium trineopentyl, cyclopentadienylzirconium trimethyl, cyclopentadienylhafnium triphenyl, cyclopentadienylhafnium trineopentyl, and cyclopentadienylhafnium trimethyl.

Other metallocenes which may be usefully employed to prepare the polymer components of the invention include the monocyclopentadienyls titanocenes such as, pentamethylcyclopentadienyl titanium trichloride,

pentaethylcyclopentadienyl titanium trichloride; bis(cyclopentadienyl)zirconium diphenyl, bis(pentamethylcyclopentadienyl) titanium diphenyl, the carbene represented by the formula bis(cyclopentadienyl)titanium=CH_2 and derivatives of this reagent such as bis(cyclopentadienyl)Ti=CH_2.Al(CH_3)$_3$, (Cp_2TiCH_2)$_2$, $Cp_2TiCH_2CH(CH_3)CH_2$, Cp_2Ti-$CHCH_2CH_2$; substituted bis(cyclopentadienyl)titanium (IV) compounds such as: bis(indenyl)titanium diphenyl or dichloride, bis(methylcyclopentadienyl)titanium diphenyl or dihalides; dialkyl, trialkyl, tetra-alkyl and penta-alkyl cyclopentadienyl titanium compounds such as bis(1,2-dimethylcyclopentadienyl)titanium diphenyl or dichloride, bis(1,2-diethylcyclopentadienyl)titanium diphenyl or dichloride and other dihalide complexes; silicon, phosphine, amine or carbon bridged cyclopentadiene complexes, such as dimethyl silyldicyclopentadienyl titanium diphenyl or dichloride, methyl phosphine dicyclopentadienyl titanium diphenyl or dichloride, methylenedicyclopentadienyl titanium diphenyl or dichloride and other dihalide complexes and the like.

Additional zirconocene catalysts useful according to the present invention include bis(cyclopentadienyl) zirconium dimethyl; bis(cyclopentadienyl) zirconium dichloride, bis(cyclopentadienyl) zirconium methylchloride, pentamethylcyclopentadienyl zirconium trichloride, pentaethylcyclopentadienyl zirconium trichloride, bis(pentamethylcyclopentadienyl)zirconium diphenyl, the alkyl substituted cyclopentadienes, such as bis(ethylcyclopentadienyl)zirconium dimethyl, bis(β-phenylpropylcyclopentadienyl)zirconium dimethyl, bis(methylcyclopentadienyl)zirconium dimethyl, bis(n-butyl-cyclopentadienyl)zirconium dimethyl, bis(cyclohexylmethylcyclopentadienyl)zirconium dimethyl, bis(n-octyl-cyclopentadienyl)zirconium dimethyl, and haloalkyl and dihalide complexes of the above; di-alkyl, trialkyl, tetra-alkyl, and penta-alkyl cyclopentadienes, such as bis(pentamethylcyclopentadienyl)zirconium

di-methyl bis(1,2-dimethylcyclopentadienyl)zirconium dimethyl and dihalide complexes of the above; silicon, phosphorus, and carbon bridged cyclopentadiene complexes such as dimethylsilyldicyclopentadienyl zirconium dimethyl or dihalide, and methylene dicyclopentadienyl zirconium dimethyl or dihalide, and methylene dicyclopentadienyl ethylene bridged bis(tetrahydroindenyl) zirconium dimethyl or dihalide, carbenes represented by the formula $Cp_2Zr=CHP(C_6H_5)_2CH_3$, and derivatives of these compounds such as $Cp_2\overline{ZrCH_2CH(CH_3)CH_2}$.

Bis(cyclopentadienyl)hafnium dichloride, bis(cyclopentadienyl)vanadium dichloride and the like are illustrative of other metallocenes.

The alumoxanes are polymeric aluminum compounds which can be represented by the general formulae $(R\text{-}Al\text{-}O)_n$ which is a cyclic compound and $R(R\text{-}Al\text{-}O\text{-})_nAlR_2$, which is a linear compound. In the general formula R is a C_1-C_5 alkyl group such as, for example, methyl, ethyl, propyl, butyl and pentyl and n is an integer from 2 to about 20. Generally, in the preparation of alumoxanes from, for example, aluminum trimethyl and water, a mixture of the linear and cyclic compounds is obtained.

The alumoxane can be prepared in various ways. Preferably, they are prepared by contacting water with a solution of aluminum trialkyl, such as, for example, aluminum trimethyl, in a suitable organic solvent such as benzene or an aliphatic hydrocarbon. For example, the aluminum alkyl is treated with water in the form of a moist solvent. In an alternative method, the aluminum alkyl such as aluminum trimethyl can be desirably contacted with a hydrated salt such as hydrated copper sulfate.

Preferably, the alumoxane is prepared in the presence of a hydrated ferrous sulfate as described in U.S. Patent 4,665,208. The method comprises treating a dilute solution of aluminum trimethyl in, for example, toluene, with ferrous sulfate represented by the general formula

$FeSO_4.7H_2O$. The ratio of ferrous sulfate to aluminum trimethyl is desirably about 1 mole of ferrous sulfate for 6 to 7 moles of aluminum trimethyl. The reaction is evidenced by the evolution of methane.

The ratio of aluminum in the alumoxane to total metal in the metallocenes can be in the range of about 0.5:1 to about 10,000:1, and preferably about 5:1 to about 1000:1.

Various inorganic oxide supports may be used for supported catalyst systems to prepare interpolymers and blend components of the present invention. The polymerization is generally carried out in the temperature range of about 0-160°C, or even higher, but this range is not meant to be exclusive for preparing the interpolymer and blend components of the invention, which may be prepared by any technique resulting in the structure set forth. Atmospheric, sub-atmospheric, or super-atmospheric pressure conditions have been used for the polymerization using the metallocene catalyst described above. It is generally preferred to use catalyst compositions at a concentration so as to provide from about 1 ppm to about 5000 ppm, most preferably 10 ppm to 300 ppm, by weight of transition metal based on the weight of monomers in the polymerization of the ethylene polymers.

A slurry polymerization process generally uses super-atmospheric pressures and temperatures in the range of 40-110°C. In a slurry polymerization, a suspension of solid, particulate polymer is formed in a liquid polymerization medium to which ethylene and comonomers and often hydrogen along with catalyst are added. The liquid employed in the polymerization medium can be alkane or cycloalkane, or an aromatic hydrocarbon such as toluene, ethylbenzene or xylene. The medium employed should be liquid under the conditions of polymerization and relatively inert. Preferably, hexane or toluene is employed.

In a modification, the polymer components of the present invention may be formed by gas-phase

polymerization. A gas-phase process utilizes super-atmospheric pressure and temperatures in the range of about 50°-120°C. Gas-phase polymerization can be performed in a stirred or fluidized bed of catalyst and product particles in a pressure vessel adapted to permit the separation of product particles from unreacted gases. Thermostated ethylene, comonomer, hydrogen and an inert diluent gas such as nitrogen can be introduced or recirculated so as to maintain the particles at a temperature of 50°-120°C. Triethylaluminum may be added as needed as a scavenger of water, oxygen, and other adventitious impurities. Polymer product can be withdrawn continuously or semi-continuously at a rate such as to maintain a constant product inventory in the reactor. After polymerization and deactivation of the catalyst, the product polymer can be recovered by any suitable means. In commercial practice, the polymer product can be recovered directly from the gas phase reactor, freed of residual monomer with a nitrogen purge, and used without further deactivation or catalyst removal. The polymer obtained can be extruded into water and cut into pellets or other suitable comminuted shapes. Pigments, antioxidants and other additives may be added to the polymer, as is known in the art.

The blends of the present invention are prepared by blending the desired components in the desired proportions using conventional blending, techniques and apparatus, such as, for example, screw-type extruders, Banbury mixers, and the like. Alternatively, the blends may be made by direct polymerization, without isolation of the blend components, using, for example, two or more catalysts in one reactor, or by using a single catalyst and two or more reactors in series or parallel. The blend may also be compounded with various conventional additives known in the art such as, for example, antioxidants, UV stabilizers, pigments, fillers, slip additives, block additives, and the like. The blend preferably does not

contain any blend components in proportions which would significantly adversely affect any improved properties desired to be obtained by blending the LLDPE resins.

The invention is illustrated by way of the following examples:

Example 1

An ethylene homopolymer resin, identified as '006, having characteristics shown in Table 1, was prepared in a gas phase reactor employing a silica supported transition metal catalyst described below.

The gas phase reaction was carried out at 185°F reaction temperature, 195 psi ethylene pressure, a hydrogen/ethylene mole ratio of 0.00041, and a residence time of 2 hours. A second polymer, an ethylene/butene-1 copolymer identified as '013, having characteristics shown in Table 1 was prepared in the gas phase reactor using the same supported catalyst as described below. The gas phase reaction was carried out at 140°F reactor temperature, 195 psi ethylene pressure, a hydrogen/ethylene mole ratio of 0.00061, a butene-1/ethylene mole ratio of 0.123, and a residence time of 2 hours.

The silica supported transition metal catalyst was obtained as follows:

100 grams of high surface area (Davison 952) silica, dehydrated in a flow of dry nitrogen at 800°C for 5 hours, was slurried with 500 cc of toluene at 25°C under nitrogen in a 2500 cc round-bottom flask using a magnetic stirrer. 250 cc of methyl alumoxane in toluene (1.03 mole/liter in aluminum) was added dropwise over 15 minutes with constant stirring to the silica slurry. Stirring was continued for 30 minutes while maintaining the temperature at 25°C. To the alumoxane treated silica slurry was added dropwise over 15 minutes, with constant stirring 250.0 cc of toluene solution containing 2.00 grams of bis(n-butylcyclopentadienyl)zirconium dichloride. This slurry was stirred an additional one half hour while maintaining the temperature at 65°C and thereafter the

toluene was decanted and the solids recovered and dried in vacuo for 4 hours. Analysis of the catalyst indicated that it contained 4.5 wt% aluminum and 0.63 wt% zirconium.

Resins '006 and '013 were then intensively mixed at 360°F in equal proportions in a Banbury melt blender along with 1000 ppm each of the stabilizers Irganox 1076 and Irgafos 168. The molten blend from the Banbury was transferred to a roll mill, made into a thin sheet and then cooled to room temperature. The cold sheet was then chopped into approximately 1/8 inch chips. Characteristics of this blend, identified as Blend B1, are given in Table 1.

Compression molded sheets, five thousandth of an inch in thickness, were prepared by melting the chopped blend at 160°C in a press, compressing to 20 tons pressure for 5 minutes and then cooling the blend under pressure to room temperature at a rate of 15°C per minute. Properties of the molded sheet specimen are shown in Table 2.

Example 2

A second compression molded sheet was prepared and tested as in Example 1, from the resins described in Example 1, except that the blend which is identified as Blend B2, consisted of one part '006 and three parts '013. Characteristics of Blend B2 are given in Table 1, properties of Blend B2 are given in Table 2.

TABLE 1
POLYMER CHARACTERIZATION

Sample Designation	Mole% Butene[a]	Mw[b]	Mw/Mn[b]	Density[c]	Melt Index[d]	CDBI[e]
'006 A	0.0%	76,700	2.8	0.9552	5.0	100%
'013 B	6.4	80,400	2.3	0.9042	4.0	67%
Blend B1	2.7	76,500	2.4	0.9335	4.4	25%
Blend B2	4.9	76,900	2.3	0.9172	4.3	44%

a. By C-13 NMR
b. Weight average molecular weight (Mw), and ratio of number average molecular weight (Mn), Mw/Mn by size exclusion chromatography.

c. Density by gradient column method, ASTM D1585, g/cm^3
d. Melt index by ASTM D1738 Condition E, dg/min
e. CDBI, composition distribution breadth index, is defined as the weight percent of polymer molecules having a comonomer content within 50% of the median total molar comonomer content. The CDBI of linear polyethylene is defined to be 100%. Determined using temperature rising elution fractionation using tetra-chloroethylene solvent over a 6°C to 120°C temperature range as described in U.S. Patent Application Serial Number 151,350. See also Wild, et al, J. Poly. Sci. Ed, Vol. 20, p 441 (1982).

TABLE 2
TEAR AND TENSILE STRENGTH OF MOLDED SHEET

Sample Designation	Tear Strength[e]	Tensile Strength[f] @ Yield	Tensile Strength[f] @ Break	Elongation[f] @ Yield	Elongation[f] @ Break	Modulus[f] Secant	Modulus[f] Tangent
'006	48	3640 psi	840	18%	710%	127,000	155,000
'013	111	800	3230	70	1830	165,000	---
Blend B1	210	1900	2870	23	1620	70,600	76,000
Blend B2	227	1330	3030	28	1530	37,700	42,000

e. Tear strength, in g/mil, measured using ASTM 1922, tear resistance by pendulum method
f. Tensile strength, elongation, and modulus measured using ASTM D638

Example 3

Polymers used as blend components were synthesized by a procedure similar to that described below for Run 03. A stainless steel, 1-liter Zipperclave autoclave reactor was used in the syntheses. The reactor was equipped with a water jacket for heating and cooling, a septum inlet for injection of liquid components, a burette for measuring and adding purified hexane, and a controlled pressure source of purified ethylene and nitrogen. Polymerization reactions were started by cleaning the reactor and purging it with purified nitrogen for several hours. 800 cc of purified hexane was added to the reactor vessel at 25°C by means of the measuring burette. 100 cc of purified 1-hexene was then added under nitrogen, and the contents of the reactor were continuously stirred at 1200rpm henceforth. 10.0 cc of the 10 wt% solution of methylalumnoxane was added by syringe, and the reactor temperature was equilibrated at the reaction temperature of 60°C. Hydrogen was not used in this example

it is added at this time in the examples which do use it. Hydrogen was metered into the reactor from a vessel of known volume equipped with a highly precise pressure gauge. The ideal gas law was used to calculate the millimoles of hydrogen introduced into the reactor. 1.00 cc of a solution of 0.01 wt% bis(n-butylcyclopentadienyl) zirconium dichloride in distilled toluene was then injected by syringe, followed by pressurization immediately with 90.9 psi of purified ethylene. Ethylene was fed continuously on demand to the stirring reactor while maintaining the reaction temperature at 60°C and the reaction pressure at 90 psi. After 10 minutes of polymerization the reaction was stopped by rapidly cooling and venting the pressure and exposing the contents to air. The polymer was obtained by evaporating the liquid components and drying in a vacuum oven for 1 hour. The poly(ethylene-co-hexene-1) polymer obtained, which is identified as CO3, was obtained in 89.7 yield.

Other polymerizations runs were performed using the procedure above with the exception that the volumes, temperatures, and other polymerization conditions were modified as shown in Table 3. Characterization data for the blend components are given in Table 4.

Blends 8A, 9D, 20C, 21F and 22E were each prepared using equal quantities of three blend components. (The letter designation in these blends corresponds to the type of blend illustrated in Figure 1.) Blend components for each of the blends are given in Table 4, and they were blended by the following solution blending procedure using ca. 14 g. of each blend component. 2000 cc of xylene was purged with nitrogen for one hour, the three blend components were added, and the mixture was stirred and heated to 144°C to dissolve the polymers. The polymer solution was then cooled to 70°C, and 2000 cc of methanol was added to precipitate the polymer. The mixture was then cooled to 22°C, and the precipitated polymer was

filtered and dried in a vacuum oven. Average composition, composition distribution breadth index, molecular weight data, and density of the blends are listed in Table 4.

Compression molded sheets, five thousandth of an inch in thickness, were prepared by melting the blended polymers in a press, compressing to 20 tons pressure for 5 minutes and then cooling the blend under pressure to room temperature. Properties of the molded sheet specimen are shown in Table 5 for blends 8A, 9D, 20C, 21F and 22E. The fraction of polymer extracted by hexane at 69°C is also given for the blends.

Table 5 shows that the tear strength of blends 9D, 20C, 21F and 22E, i.e., blends having novel composition-molecular weight distributions, are each superior to that of blend 8A, which has a conventional-type composition-molecular weight distribution. In many applications it is desirable to have low levels of extractable polymer, and the table shows that blends 9D, 20C, 21F, and 22E are markedly superior to blend 8A in this respect. Moreover, tensile properties such as tensile strength at yield of the conventional-type blend 8A are generally comparable to the tensile strengths of the other blends. Consequently, the benefits of high tear strength and low levels of extractables can be achieved without undue loss of tensile properties.

This example illustrates the benefits obtained by certain novel composition-molecular weight distributions differing from the conventional type. Thus, blends 9D and 20C have similar average molecular weights and molecular weight distributions. Consequently, flow and melt fabrication performance of these blends should be similar to that of the conventional-type blend 8A. Therefore, the desirable mechanical and solubility properties of blends 9D and 20C are not obtained at the expense of fabrication performance. Additionally, blends 21F and 22E have much lower weight average molecular weight than blend 8A, and

they should therefore be more easily fabricated into articles of commerce such as films, fibers and molded objects.

In summary, in the blends of this example (Type C, D, E and F blends) the average molecular weight and molecular weight distribution can be manipulated independently of the composition distribution. Superior solid state performance can therefore be achieved without sacrificing melt fabrication performance.

TABLE 3
POLYMERIZATION CONDITIONS

Polymerization Run No.	Reaction Temp.°C	$C_2^=$ Press. (psi)	Time (min.)	Hexane (cc)	1-Hexene (cc)	Catalyst Solu. (cc)	Yield gm	H_2 m mole
03	60	90	10	800	100	1.0	89.7	--
09	80	60	10	800	75	1.0	36.6	--
15	80	60	15	800	--	1.0	47.0	10
19	80	60	60	800	75	0.25	44.2	5
27	90	60	45	800	150	0.50	96.5	--
30	75	75	42	800	87.5	0.55	86.3	--
38	60	130	20	800	150	0.20	89.5	--
42	60	60	30	800	--	1.0	81.0	--
48	70	170	9	300	600	0.5	114.0	--
64	68	302	60	--	800	0.05	53.0	--

TABLE 4
POLYMER CHARACTERIZATION OF BLENDS AND BLEND COMPONENTS

Sample[h]	Mole %[a] Hexene	Mw[b]	Mw/Mn	Density[b]	CDBI[b]
C27	6.1	12,500	1.9	--	77%
C30	2.0	48,800	1.9	--	81
C42	0.0	259,000	2.0	--	100
Blend 8A[c]	2.7	113,000	9.0	0.933	24
C15	0.0	13,400	2.7	--	100
C30	2.0	48,000	1.9	--	81
C64	4.2	223,000	2.2	--	--
Blend 9D[d]	2.1	95,600	9.8	0.940	42
C19	2.3	13,600	1.9	--	70
C30	2.0	48,800	1.9	--	81
C38	2.2	240,000	2.2	--	--
Blend 20C[e]	2.3	94,800	8.8	0.932	64
C15	0.0	13,400	2.7	--	100
C09	3.0	52,000	2.4	--	79
C03	3.1	121,000	2.2	--	67
Blend 21F[f]	--	62,000	6.4	0.946	46
C15	0.0	13,400	2.7	--	100
C09	3.0	52,000	2.4	--	79
C48	7.2	50,000	1.9	--	75
Blend 22E[g]	--	37,800	4.2	0.940	37

(a) Calculated from absorbance of 1378 cm^{-1} IR band using spectral substraction to correct for overlap of adjacent IR bands. Residual peak at 1378 cm^{-1} was converted to hexene content using calibration obtained by C13 NMR analysis of poly(ethylene-co-hexenes).

(b) Same as in Table 1

(c) Blend 8A consisted of equal quantities of C27, C30 and C42

(d) Blend 9D consisted of equal quantities of C15, C30 and C64

(e) Blend 20C consisted of equal quantities of C19, C30 and C38

(f) Blend 21F consisted of equal quantities of C15, C09 and C03

(g) Blend 22E consisted of equal quantities of C15, C09 and C48

(h) Blend components in this table are identified by the letter C followed by the corresponding polymerization run number from Table 3. For example, blend component C27 was obtained from polymerization run 27.

TABLE 5
PROPERTIES OF MOLDED SHEET

Sample	Tear Strength g/mil	Tensile Strength at Yield, psi	Hexane Extractable Polymer %
Blend 8A	68	1770	22
Blend 9D	241	1980	3.0
Blend 20C	426	1400	6.3
Blend 21F	205	2050	4.5
Blend 22E	107	2050	6.7

(a) Tear strength measured by ASTM 1922, tear resistance by pendulum method.
(b) Tensile strength at yield measured using ASTM D638.
(c) Extractables were determined as follows. About 0.5 g of polymer molded sheet, ca. 5 mil thick, was weighed and placed in a weighed cellulose extraction thimble. The thimble and polymer were placed in a vapor jacketed Soxlet-type extractor, and the polymer was extracted with condensed vapors of boiling hexane at 69°C for 16 hours. The thimble and residual polymer were removed, dried in a vacuum oven, and weighed to determine the amount of extracted, i.e., soluble, polymer.

The foregoing description of the invention is illustrative and explanatory thereof. Various changes in the materials, apparatus, and particular steps employed will occur to those skilled in the art. It is intended that all such variations within the scope and spirit of the appended claims be embraced thereby.

CLAIMS:

1. A polymer blend, comprising:

a plurality of linear ethylene interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%;

wherein the blend is essentially free of blend components having both a higher average molecular weight and a lower average comonomer content than that of any other said polyethylene blend component; and

wherein the blend has either $M_w/M_n > 3$ or a composition distribution breadth index below 50%, or both.

2. The blend of claim 1, wherein said blend has a density in the range of 0.85 to 0.94 g/cm^3.

3. The blend of claim 1, wherein said blend components have an α-olefin comonomer content in the range of 0 to 30 mole percent and an ethylene content in the range of 100 to 70 mole percent.

4. The blend of claim 1, wherein said blend components are selected from the group consisting of:

poly(ethylene-co-α-olefin) elastomers; very low density linear polyethylenes; and linear, low density polyethylenes.

5. The blend of claim 1, wherein the blend components have essentially the same molecular weight and different comonomer contents.

6. The blend of claim 1, wherein the blend components have essentially the same comonomer content and different molecular weights.

7. The blend of claim 1, wherein the molecular weights and the comonomer contents of the blend components are both different.

8. The blend of claim 1, wherein the value of M_w/M_n for each blend component is less than or equal to 2.5.

9. The blend of claim 1, wherein the composition distribution breadth index for each blend component is greater than about 70%.

10. A polymer blend, comprising:

a plurality of linear ethylene polymer or interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%; and

wherein said blend components are selected from one of the groups consisting of: (1) blend components having essentially the same average molecular weight but different average comonomer contents; (2) blend components having essentially the same average comonomer content but different average molecular weights; and (3) blend components having different average molecular weights and comonomer contents wherein said components, taken serially in order of increasing average molecular weight, have an increasing comonomer content.

11. The blend of claim 10, wherein the blend has a density of 0.85 to 0.94 g/cm^3.

12. The blend of claim 10, wherein said blend has an average molar α-olefin comonomer content of 1-30 percent.

13. The blend of claim 10, wherein said blend components include poly(ethylene-co-α-olefin) elastomers.

14. The blend of claim 12, wherein said blend components include very low density, linear polyethylenes.

15. The blend of claim 12, wherein said blend components include linear, low density polyethylenes.

16. The blend of claim 10, wherein the number of said blend components is from 2 to 5.

17. The blend of claim 10, wherein said blend is essentially free of blend components having both a higher average molecular and a lower average comonomer content than another blend component.

18. The blend of claim 10, wherein the blend has plural modality with respect to molecular weight, comonomer content or both.

19. The blend of claim 12, wherein said α-olefins have from 3 to about 12 carbon atoms.

20. The blend of claim 10, wherein $M_w/M_n \leq 2.5$ for each blend component, and the composition distribution breadth index for each component is in the range from 50 to 70%.

21. The blend of claim 20, wherein $M_w/M_n \leq 2.0$ for each blend component.

22. The blend of claim 20, wherein each blend component has a composition distribution breadth index in the range from 50 to 90%.

23. A polymer blend exhibiting enhanced heat strength properties over that of the individual blend components comprising:

a plurality of linear ethylene interpolymer blend components, each having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ and a composition distribution breadth index of at least 50%; and

wherein for said components taken serially, primarily in order of increasing molecular weight and secondarily in order of increasing average comonomer content, each succeeding component has substantially the same or a higher average comonomer content than each preceding component in said series.

24. A polymer blend of linear ethylene interpolymer having a molecular weight distribution such that $M_w/M_n > 2$ and a composition distribution breadth index greater than or equal to 50%.

25. A linear ethylene interpolymer comprising fractions having a narrow molecular weight distribution such that $M_w/M_n \leq 3$ for each fraction, said fractions taken serially in order of increasing average molecular weight have an increasing average comonomer content.

26. A linear ethylene interpolymer comprising fractions having a composition distribution breadth index of 50% or more, said fractions taken serially in order of increasing comonomer content have an increasing average molecular weight.



Gp 1505

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Pak-Wing Steve Chum et al.

Serial No.: 08/054,379 Group Art Unit: 1505

Filed: April 28, 1993 Examiner: D. WU

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO COMMISSIONER OF PATENTS AND TRADEMARKS, WASHINGTON, D.C. 20231, On May 13, 1994
DATE OF DEPOSIT
Jan Alverson
PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE
Jan Alverson
SIGNATURE OF PERSON SIGNING CERTIFICATE
5-13-94
DATE OF SIGNATURE

RECEIVED
JUN 01 1994
GROUP 150

Hon. Commissioner of Patents & Trademarks
Washington, D.C. 20231

Sir:

EXTENSION OF TIME

Applicant(s) hereby request(s) that the shortened statutory period for response now set to expire February 15, 1994 in this application be extended for a period of three (3) months, the extended period then expiring on May 15, 1994. This is a first request for an extension of time.

Please charge $840 to our Deposit Account No. 04-1512. If this estimate is incorrect, please charge or credit our account accordingly. Two duplicate copies of this sheet are enclosed.

Respectfully submitted,

By Noreen D Warrick
Noreen D. Warrick
Registration No. 34,573
Phone: 409-238-4510

May 11, 1994
Freeport, Texas 77541

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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Pak-Wing Steve Chum et al.

Serial No.: 08/054,379

Filed: April 28, 1993

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

Docket No.: C-40,121-G

Group Art Unit: 1505

Examiner: D. Wu

RECEIVED
JUN 0 1 1994
GROUP 150

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Commissioner of Patents and Trademarks
Washington, D.C. 20231

Sir:

RESPONSE A

In response to the Office Action dated November 15, 1993, Applicants have enclosed a request for a three (3) month extension of time, thereby extending the period of response to May 15, 1994. In addition, Applicants respectfully request entry of the following amendments and reconsideration in view of the following remarks:

In the Specification:

Page 1, line 3, after the words "number 07/776,130," please insert the words -- now USP 5,272,236, -- ;

p. 1, line 4, after the words "number 07/939,281," please insert the words -- now USP 5,278,272, -- ;

p. 3, line 2, please delete the word "temperture" and insert therefore the word -- temperature --;

p. 3, line 14, please the letters "e.g," and insert therefore the letters -- e.g., --;

p. 3, line 15, after the words "The compositions comprise" please insert the letter -- (A) --;

p. 3, line 17, please delete the symbol " : ";

p. 3, line 18, please delete the letter "(A)";

p. 5, line 9, please delete the word "homogeniety" and insert therefore the word -- homogeneity --;

p. 5, line 28, please delete the word "homogeneouslt" and insert therefore the word -- homogeneously --;

p. 8, line 4, please delete the word "substantialy" and insert therefore the word -- substantially --;

p. 10, lines 11 and 16, please delete the word "intial" and insert therefore the word -- initial --;

p. 11, line 2, please delete the word "heterogenously" and insert therefore the word -- heterogeneously --;

p. 14, line 5, please delete the word "tempertaures" and insert therefore the word -- temperatures --;

p. 14, line 9, please delete the word "concommitantly" and insert therefore the word -- concomitantly --;

p. 17, line 23, please delete the word "decribed" and insert therefore the word -- described --;

p. 17, line 27, please delete the word "dicusses" and insert therefore the word -- discusses --;

p. 18, line 11, please delete the word "terepthalate" and insert therefore the word -- terephthalate --;

p. 22, line 5, please delete the word "coplymer" and insert therefore the word -- copolymer --;

p. 22, line 10, please delete the word "recator" and insert therefore the word -- reactor --;

p. 23, line 4, please delete the word "baout" and insert therefore the word -- about --;

p. 23, line 5, please delete the word "distrubution" and insert therefore the word -- distribution --;

p. 26, line 11, please delete the word "ipact" and insert therefore the word -- impact --;

p. 26, line 12, please delete the words "DC ppt tear" and insert therefore the words -- CD ppt tear --.

In the Claims:

Please cancel Claims 9-16 and 24-30 without prejudice.

Please amend the following claims:

1. (Amended) A film made from an ethylene polymer composition, wherein the composition comprises

(A) from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of[:]

[(A)] at least one homogeneously branched substantially linear ethylene/α-olefin interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.92 g/cm^3,

(ii) a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8,

(iii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min) to about 10 g/10 min,

(iv) no linear polymer fraction, and

(v) a single melting peak as measured using differential scanning calorimetry; and

(B) from about 5 percent (by weight of the total composition) to about 90 percent (by weight of the total composition) of at least one heterogeneously branched ethylene polymer having a density from about 0.93 g/cm^3 to about 0.965 g/cm^3 and wherein the heterogeneously branched ethylene polymer has no more than about

10 percent (by weight of the polymer) of a polymer fraction having a slope of strain hardening coefficient (SHC) greater than or equal to about 1.3.

2. (Amended) The film of claim 1 wherein the homogeneously branched substantially linear ethylene/α-olefin interpolymer has a slope of strain hardening coefficient greater than or equal to about 1.3 and up to about 2.3.

---

17. (Amended) In a composition comprising at least one homogeneously branched ethylene/α-olefin interpolymer and at least one heterogeneously branched ethylene/α-olefin interpolymer, the improvement comprising incorporating into the composition from about 10 percent (by weight of the total composition) to about 95 percent (by weight of the total composition) of at least one homogeneously branched substantially linear ethylene/α-olefin interpolymer having:

(i) a density from about 0.89 grams/cubic centimeter (g/cm^3) to about 0.92 g/cm^3,

(ii) a molecular weight distribution (M_w/M_n) from about 1.8 to about 2.8,

(iii) a melt index (I_2) from about 0.001 grams/10 minutes (g/10 min) to about 10 g/10 min,

(iv) no linear polymer fraction, and

(v) a single melting peak as measured using differential scanning calorimetry,

wherein the heterogeneously branched ethylene polymer has no more than about 10 percent (by weight of the polymer) of a polymer fraction having a slope of strain hardening coefficient greater than or equal to about 1.3.

18. (Amended) The improvement of claim 17 wherein the homogeneously branched substantially linear ethylene/α-olefin interpolymer has a slope of strain hardening coefficient greater than or equal to about 1.3 and up to about 2.3.

REMARKS

The amendments to the specification update the priority application data and correct obvious typographical spelling errors. Applicants regret the errors and respectfully request that the amendments to the specification be entered.

The amendments to independent Claims 1 and 17 describing the heterogeneously branched ethylene polymer are supported in the specification on p. 14, lines 22-23. The other amendment to independent Claim 1 clarifies the composition used to make the film as comprising components (A) and (B). The amendments to dependent Claims 2 and 18 are supported in Table 1, p. 12, in Polymer Q. These amendments do not add new matter. Entry of the amendments is respectfully requested.

Applicants also affirm the election of Group I, Claims 1-8 and 17-23. Applicants reserve the right to timely file divisional applications to the non-elected subject matter of Group II (Claims 9-16 and 24-30).

THE REJECTIONS

1. Claims 1-8 and 17-23 are rejected under 35 U.S.C. § 112, second paragraph, as allegedly being indefinite for failing to particularly point out and distinctly claim the subject matter. Specifically, the language "substantially linear ethylene/alpha-olefin interpolymer" in claims 1 and 17 is allegedly in conflict with language "no linear polymer fraction."

2. Claims 2 and 18 are rejected under 35 USC 112, first paragraph, as the disclosure is allegedly enabling only for claims limiting the slope of strain hardening coefficient up to 2.265.

3. Claims 1-8 and 17-23 are rejected under 35 USC 102(b) as anticipated by, or in the alternative, under 35 USC 103 as obvious over, WO 90/03414 (WO '414).

4. Claims 1-8 and 17-23 are rejected under 35 USC 103 as being unpatentable over USP 5,206,075 (Hodgson).

APPLICANTS' RESPONSE

Applicants discovered that films made from specific ethylene polymer compositions have enhanced properties. The ethylene polymer compositions comprise at least one substantially linear ethylene polymer and at least one heterogeneously branched ethylene polymer. The substantially linear ethylene polymer is not a linear polymer, but has long chain branching and an absence of a linear polymer fraction. The substantially linear ethylene polymers used herein are homogeneous polymers having a narrow molecular weight distribution (MWD) and a narrow or uniform composition distribution (CD). The narrow MWD (generally, MWD ≈ 2) means that the length of the polymer chains is relatively uniform, and the narrow CD means that each polymer chain contains a relatively uniform amount (e.g. weight percent) of α-olefin in copolymerized form and randomly dispersed along the polymer backbone and along any long chain branches. The substantially linear ethylene polymer also has a uniform (i.e., homogeneous) short chain branching distribution, meaning that essentially each polymer molecule has the same number of short chain branches along the polymer chain. In contrast, the heterogeneously branched ethylene polymers, by definition, have broader MWD's, broader CD and different amounts of short chain branching. The heterogeneously branched ethylene polymers are typically prepared using Ziegler catalysis. As Figure 1 demonstrates, each of the polymer components used in Applicants' invention has different branching distributions. The novel films which Applicants have discovered have improved impact and tensile properties and an especially good combination of modulus, yield, ultimate tensile and toughness. See, for example, p. 3, lines 9-14 of the specification and Table 3 (p. 25) and the summary discussion on p. 26.

1. Applicants respectfully submit that the basis of rejection of Claims 1-8 and 17-23 under 35 U.S.C. § 112, second paragraph, is in error. Applicants have taught and claimed that the substantially linear ethylene polymer has long chain branching and that such a substantially linear ethylene polymer has no measurable linear polymer fraction (i.e., no high density fraction) as measured by temperature

rising elution fractionation. See the teaching, for example, at p. 5, lines 23-32 in the Specification. This "high density fraction" or linear polymer fraction is lacking from the substantially linear ethylene polymers used in Applicants' invention. Applicants have taught that conventional ethylene polymers (i.e., LLDPE made using Ziegler catalysis) comprise a variety of polymer molecule components, some molecules having numerous short chain branches and some having no short chain branches. The short chain branches are the result of uneven comonomer incorporation during polymerization, which results in a polymer product having several different types of polymer molecules. For example, see the discussion at p. 13, line 7 through p. 14, line 10. Figure 2 also graphically depicts the relative amounts of the short chain branching contained in a conventional Ziegler polymerized polyethylene polymer (Dowlex™ 2045, a heterogeneously branched ethylene/1-octene copolymer made by The Dow Chemical Company). Note how the ATREF curve is much broader over the elution temperature range for the Dowlex™ 2045 polymer, as compared to the homogeneously branched substantially linear ethylene polymers. Applicants respectfully believe that there is no confusion by using the terms "substantially linear ethylene/alpha-olefin interpolymer" and "no linear polymer fraction" to describe the properties of the substantially linear ethylene polymer, since such terms are not contradictory, but rather complimentary. Accordingly, Applicants respectfully traverse the rejection of claims 1-8 and 17-23 under 35 U.S.C. § 112, second paragraph as being indefinite.

2. Applicants have listed substantially linear ethylene polymers with their respective slope of strain hardening coefficients in Table 1. The data of Table 1 is also plotted in Figure 1. Applicants have shown that a substantially linear ethylene polymer having a density of about 0.9216 g/cm^3 has a slope of strain hardening coefficient (SHC) of about 2.3 (see Polymer Q of Table 1). Applicants have amended dependent claims 2 and 18 to incorporate this upper limit for SHC and respectfully traverse the rejection of claims 2 and 18, as amended, under 35 USC 112, first paragraph.

3. WO '414 discloses blends of ethylene interpolymer components with narrow molecular weight and composition

distributions. Each component of WO '414 has a M_w/M_n value less than or equal to 3 and a composition distribution breadth index (CBDI) of 50% or higher. Furthermore, the blend components are each linear. See page 6, line 14, 7, line 28, and page 12, line 33, among others. In contrast to WO '414, Applicants' film is made from ethylene polymer compositions which comprise at least one substantially linear ethylene polymer and at least one heterogeneously branched ethylene polymer (i.e., a conventional LLDPE, as WO '414 describes such heterogeneously branched ethylene polymers). The substantially linear ethylene polymer used in Applicants' invention is not a linear polymer, but instead is an ethylene polymer having long chain branching. The substantially linear ethylene polymers used in Applicants' invention are patentably distinct from the linear ethylene polymers used in WO '414 (see USP 5,272,236 and USP 5,278,272). In addition, the heterogeneously branched ethylene polymer used as the other component of the ethylene polymer compositions used to make the novel film has a broad short chain branching distribution, as Figure 2 demonstrates. WO '414 specifically calls for their blends to each have a CBDI of at least 50%. Consequently, both components of the film of Applicants' ethylene polymer compositions are different from the homogeneously branched linear ethylene polymers used in the blends of WO '414.

WO '414 neither teaches nor suggests the use of a substantially linear ethylene polymer in combination with a heterogeneously branched ethylene polymer. However, WO '414 does lead one skilled in the art away from the use of conventional heterogeneously branched ethylene polymers in their blends. For example, WO '414 states: "The higher molecular weight fraction containing relatively less comonomer as on conventional LLDPE may cause an anisotropic morphology during fabrication processing known as 'row nucleated' or 'shish-ka-bob' morphology. *This anisotropic morphology is believed to contribute to poor toughness in articles crystallized from flowing melts.*" (Page 11, lines 14-20, emphasis added in italics.).

Thus, in direct contrast to that discovered by Applicants, WO '414 concludes that blends of a homogeneously branched ethylene polymer with conventional LLDPE results in thermally formed articles having

poor toughness. Applicants have shown in Table 3 that Examples 1 and 2 both have significantly higher dart impact than comparative example 3. Examples 1 and 2 also have higher machine and cross direction toughness than comparative example 3.

WO '414 specifically describe their polymer components as linear and as having CBDI greater than 50%, and also teach against using conventional (i.e., Ziegler polymerized heterogeneously branched) LLDPE. See also Figure 2 of WO '414, showing the broad CBDI of a "typical prior art LLDPE." (p. 5, lines 28-30 and Figure 2 of WO '414). Accordingly, WO '414 neither anticipates nor obviates Applicants' claimed invention of film made from an ethylene polymer composition comprising at least one substantially linear ethylene polymer and at least one heterogeneously branched ethylene polymer. Thus, Applicants respectfully traverse the rejection of claims 1-8 and 17-23 under 35 USC 102(b) as anticipated by, or in the alternative, under 35 USC 103 as obvious over WO '414.

4. Hodgson discloses film made from a base layer. The base layer has a heat sealable film layer on one or both surfaces of the base layer. The base layer comprises a blend of at least one very low density ethylene copolymer having a CBDI of 70% or higher and another polymer. The other polymer of the base layer is selected from the group consisting of polypropylene, low density polyethylene, linear low density polyethylene (LLDPE), polybutene, random copolymers of propylene with up to about 15 mole % of a C_2 or C_4 to C_{12} alpha olefin (see col. 4, lines 14-20 of Hodgson). However, Hodgson does not specify or describe the linear low density polyethylene as the "other polymer" of the base layer. Hodgson neither teaches nor suggests that the LLDPE which can be added to his film compositions is a heterogeneously branched polyethylene, as Applicants have taught and claimed.

Furthermore, as Inventor Chum's attached declaration shows, not all heterogeneously branched linear low density polyethylene has no more than about 10 % of a polymer fraction having a slope of strain hardening coefficient (SHC) greater than about 1.3. Inventor Chum points out that about 30 % of Dowlex™ 2045 (a heterogeneously branched ethylene/1-octene copolymer made by The Dow Chemical Company having a whole polymer density of about 0.92 g/cm^3) has

SHC > 1.3. Please note that the specification on p. 13, lines 26-31 describes Dowlex™ 2045 and that Figure 2 of the instant application shows its branching distribution. Thus not all heterogeneously branched ethylene polymers will have the properties necessary for improving the film performance.

Furthermore, the other component of the compositions used in Hodgson (as well as the other layer of the film) is "VLDPE." VLDPE is well known in the art to refer to a linear ethylene polymer having a very low density. Very low density polyethylene and ultra low density polyethylene (ULDPE) are used interchangeably in the art and refer to linear polyethylenes having densities less than 0.915 g/cm^3. *See* L.D. Cady, *Ultralow-density Polyethylene*, Modern Plastics Encyclopedia/89, mid October 1988 Issue, Volume 65, Number 11, p. 66 (copy enclosed). The substantially linear ethylene polymers used in Applicants' invention are patentably distinct from such VLDPE polymers, as discussed supra.

Applicants' invention is directed to film made from at least one substantially linear ethylene polymer and at least one heterogeneously branched ethylene polymer having not more than 10% of a polymer fraction having SHC ≥ 1.3. Since Hodgson is directed to linear ethylene polymers blended with some unknown and unspecified type of LLDPE, Applicants believe that Hodgson does not obviate the presently claimed invention. Accordingly, Applicants respectfully traverse the rejection of claims 1-8 and 17-23, as amended, under 35 USC 103 as being unpatentable over USP 5,206,075 (Hodgson).

CONCLUSION

Applicants respectfully submit that all of the rejections have been traversed. The claims, as amended, are believed to be in condition for allowance. Applicants respectfully request reconsideration and notice of allowance.

Respectfully submitted,

Noreen D Warrick

Noreen D. Warrick
Registration No. 34,573
Telephone: (409) 238-4510

May 12, 1994

The Dow Chemical Company
Patent Department
Building B-1211
Freeport, Texas 77541



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Pak-Wing Steve Chum et al.

Serial No.: 08/054,379 Group Art Unit: 1505

Filed: April 28, 1993 Examiner: D. WU

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO COMMISSIONER OF PATENTS AND TRADEMARKS, WASHINGTON D.C. 20231, On May 13, 1994

DATE OF DEPOSIT

Jan Alverson

PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

Jan Alverson

SIGNATURE OF PERSON SIGNING CERTIFICATE

5-13-94

DATE OF SIGNATURE

RECEIVED

JUN 0 1 1994

GROUP 150

Hon. Commissioner of Patents & Trademarks
Washington, D.C. 20231

Sir:

DECLARATION UNDER CFR 1.132

Pak-Wing Steve Chum declares and states:

THAT, he obtained a Bachelor of Science degree in Chemistry from Hong Kong Baptist College in 1970, a Master of Science degree in Chemistry from Northern Michigan University in 1974, and a PhD in Physical Organic Chemistry from Oregon State University in 1978;

THAT, he joined The Dow Chemical Company (Midland, MI) in 1980 as a Senior Research Chemist, was promoted to Project Leader in 1983, to Research Leader in 1985, to Research Associate in 1988, to Associate Scientist in 1991, and to his present position as Senior Associate Scientist in 1994;

THAT, during the last fourteen years, he has focused his research effort in the area of thermolplastics and plastic foams;

THAT, he currently leads the effort in Polymer Materials Science research for The Dow Chemical Company (Texas Operations);

That, he is a member of the American Chemical Society, a member of the Board of Directors of the Society of Plastic Engineers, a member of the Board of Industry Advisors for Case Western Reserve University, and was a Visiting Professor for the Michigan Macromolecular Institute (1987-1988);

THAT, he received the American Chemical Society Outstanding Scientific Achievement Award in 1993;

THAT, he served as author or co-author of the publications 1992-1994 listed in Attachment One;

That, he is an inventor in the above-identified Application and is therefore familiar with the Application and the Office Action thereon;

THAT, polyethylenes, including linear low density polyethylene, can be characterized by various slope of strain hardening coefficient profiles;

THAT, not all linear low density polyethylene is characterized as having no more than about 10 percent of a polymer fraction having a slope of strain hardening coefficient greater than about 1.3;

THAT, for example, about 30 percent of DOWLEX™ 2045 (a heterogeneously branched linear low density ethylene/1-octene copolymer made by The Dow Chemical Company having a whole polymer density of about 0.92 g/cm³) is characterized by a slope of strain hardening coefficient greater than about 1.3;

THAT, films fabricated from polymer blends with compositions comprising: (a) a heterogeneously branched ethylene polymer having a density from about 0.93 to 0.965 g/cm³ and are characterized as having no more than about 10 percent of a polymer fraction having a slope of strain hardening coefficient greater than about 1.3, and (b) a homogeneously branched substantially linear ethylene/α-olefin interpolymer, are improved over films fabricated from conventional linear low density polyethylene, such as are prepared using Ziegler-Natta catalysts comprising a heterogeneously branched ethylene polymer characterized as having more than about 10 percent of a polymer fraction having a slope of strain hardening coefficient greater than about 1.3, in that the former are characterized by greatly improved dart impact A values (see Table 3 at page 25 of the specification).

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date May 12, 1994

Pak-Wing Steve Chum

PUBLICATIONS BY PAK-WING STEVE CHUM
(1992 to 1994)

1. "Computer Modeling of (110) Adjacent Re-entry of Polyethylene Molecules"; P. S. Chum (Dow Chemical Co.) and Prof. P. J. Phillips (University of Tennessee), et al., *Macromolecules, Vol. 27, No. 3, pp. 656, (1994)*

2. "Effect of Chain Microstructure on Modulus of Ethylene-alpha-Olefin Copolymers"; P. S. Chum, K. Sehanobish, C. I. Kao, et al., (all of Dow Chemical Company), *Journal of Applied Polymer Science, Vol. 51, pp. 887 (1994)*

3. "New Polyolefin Based Elatomers for TPO Impact Modification", P. S. Chum, H. C. Silvis C. I. Kao, (all of Dow Chemical Company), *Proceedings of the SAE International Congress & Exposition, Paper # 940435, February, 1994*

4. "Morphology and Deformation Behavior of Homogeneous Polyolefin Copolymers made with Insite* Technology", P. S. Chum, R. Guerra, et al., (All of Dow Chemical Co.), *Proceedings of SPE Antec' 94, May, 1994*

5. "Bridging the Polyethylene Lifetime under Fatigue and Creep Conditions with its Crystallization Behavior", P. S. Chum (Dow Chemical Company) and Prof. A. Chudnovsky (University of Illinois at Chicago), et al., *Journal of Applied Polymer Science, Vol 49, pp. 863-875 (1993)*

6. "Synthesis of a Stable Foamable, Gold-Containing Organic Carboxylate Polyethylene Polymer"; P. S. Chum (Dow Chemical Co.), Prof. J. P. Fackler (Texas A&M University), et al., *Journal of Polymer Science: Part A: Polymer Chemistry, Vol 31, PP. 2353 (1993)*

7. "Insite* Technology Based Polyolefin Elatomers for Impact Modification", P. S. Chum, H. C. Silvis and C. I. Kao (All of Dow Chemical Company), *Proceedings of the 1993 SPO International Polymer Conference, PP. 247 (1993)*

8. "Effect of Vinyl Chain-Ends on the Melt Viscoelatics Properties of Radiation Crosslinked Polyethylenes", P. S. Chum (Dow Chemical Company), *Journal of Plastics Film and Sheeting, Vol. 8, No. 1, PP. 37, (1992)*

9. "Influence of Extruder Conditions on Orientation Distribution in LLDPE Blown Films", P. S. Chum and S. V. Karande (All of Dow Chemical Co.), *Journal of Plastics Film and Sheeting, Vol. 8, No. 1, PP. 74, (1992)*

ATTACHMENT ONE

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office

Address : COMMISSIONER OF PATENTS AND TRADEMARKS
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.
08/054,379	04/28/93	CHUM	P C401216

WU, D EXAMINER

15M2/0727

THE DOW CHEMICAL CO.
PATENT DEPARTMENT
BLDG. B-1211
DALLAS, TX 77541

ART UNIT	PAPER NUMBER
1505	7

DATE MAILED: 07/27/94

This is a communication from the examiner in charge of your application.
COMMISSIONER OF PATENTS AND TRADEMARKS

☒ This application has been examined ☒ Responsive to communication filed on 5-18-94 ☐ This action is made final.

A shortened statutory period for response to this action is set to expire 3 month(s), 0 days from the date of this letter.
Failure to respond within the period for response will cause the application to become abandoned. 35 U.S.C. 133

Part I THE FOLLOWING ATTACHMENT(S) ARE PART OF THIS ACTION:

1. ☐ Notice of References Cited by Examiner, PTO-892.
2. ☐ Notice re Patent Drawing, PTO-948.
3. ☐ Notice of Art Cited by Applicant, PTO-1449.
4. ☐ Notice of informal Patent Application, Form PTO-152.
5. ☐ Information on How to Effect Drawing Changes, PTO-1474.
6. ☐ _____

Part II SUMMARY OF ACTION

1. ☒ Claims 1-8, and 17-23 are pending in the application.

   Of the above, claims _____ are withdrawn from consideration.

2. ☒ Claims 9-16 and 24-30 have been cancelled.

3. ☐ Claims _____ are allowed.

4. ☒ Claims 1-8 and 17-23 remain are rejected.

5. ☐ Claims _____ are objected to.

6. ☐ Claims _____ are subject to restriction or election requirement.

7. ☐ This application has been filed with informal drawings under 37 C.F.R. 1.85 which are acceptable for examination purposes.

8. ☐ Formal drawings are required in response to this Office action.

9. ☐ The corrected or substitute drawings have been received on _____. Under 37 C.F.R. 1.84 these drawings are ☐ acceptable. ☐ not acceptable (see explanation or Notice re Patent Drawing, PTO-948).

10. ☐ The proposed additional or substitute sheet(s) of drawings, filed on _____ has (have) been ☐ approved by the examiner. ☐ disapproved by the examiner (see explanation).

11. ☐ The proposed drawing correction, filed on _____, has been ☐ approved. ☐ disapproved (see explanation).

12. ☐ Acknowledgment is made of the claim for priority under U.S.C. 119. The certified copy has ☐ been received ☐ not been received ☐ been filed in parent application, serial no. _____; filed on _____.

13. ☐ Since this application appears to be in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under Ex parte Quayle, 1935 C.D. 11; 453 O.G. 213.

14. ☐ Other

EXAMINER'S ACTION

PTOL-326 (Rev. 9-89)

15. Applicant's election without traverse of Group I, claims 1-8, and 17-23 in Paper No. 6 is acknowledged.

16. The following is a quotation of the appropriate paragraphs of 35 U.S.C. § 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless --
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

17. The following is a quotation of 35 U.S.C. § 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.
>
> Subject matter developed by another person, which qualifies as prior art only under subsection (f) or (g) of section 102 of this title, shall not preclude patentability under this section where the subject matter and the claimed invention were, at the time the invention was made, owned by the same person or subject to an obligation of assignment to the same person.

18. Claims 1-8, and 17-23 remain rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103 as obvious over WO '414.

19. Paragraph 27 of Paper No. 4 are incorporated herein by reference.

20. The applicants argue that part (A) of claimed polymer blends is a substantially linear ethylene/alpha-olefin interpolymer which is not a linear polymer but has "long chain branching", and part (B) is a heterogeneously branched ethylene polymer wherein less than 10 wt.% of a polymer fraction having SHC > about 1.3. However, the physical properties of '006 and '013 of WO '414 such as density, MWD, melt index are substantially similar to those set forth in the claims. Moreover, the term "long chain branching" which applicants alleged as novelty of part (A) has never been set forth in the claims; the part (B) is made by a conventional Ziegler-type catalyst which is the same method '013 of WO '414 was made. Applicants also alleged that in Table 3, example 1 and 2 both have significantly higher dart impact and toughness than those of comparative example 3. However, none of the comparative experiments (including comparative example 3) are truly representative of the closest disclosure of WO '414.

The Chum's Declaration filed on May 18, 1994 has been considered but not deemed to be persuasive since there is no any comparative tests to show the unexpected results of the claimed polymer blends, mere conclusory statements are not entitled to

probative weight. Since applicants have not met their burden to provide objective evidence demonstrating the claimed polymer blends are in fact differ from those of WO '414, the 102(b)/103 rejection is still deemed to be proper.

21. Applicant's arguments filed on May 18, 1994 have been fully considered but they are not deemed to be persuasive.

22. **THIS ACTION IS MADE FINAL.** Applicant is reminded of the extension of time policy as set forth in 37 C.F.R. § 1.136(a).

A SHORTENED STATUTORY PERIOD FOR RESPONSE TO THIS FINAL ACTION IS SET TO EXPIRE THREE MONTHS FROM THE DATE OF THIS ACTION. IN THE EVENT A FIRST RESPONSE IS FILED WITHIN TWO MONTHS OF THE MAILING DATE OF THIS FINAL ACTION AND THE ADVISORY ACTION IS NOT MAILED UNTIL AFTER THE END OF THE THREE-MONTH SHORTENED STATUTORY PERIOD, THEN THE SHORTENED STATUTORY PERIOD WILL EXPIRE ON THE DATE THE ADVISORY ACTION IS MAILED, AND ANY EXTENSION FEE PURSUANT TO 37 C.F.R. § 1.136(a) WILL BE CALCULATED FROM THE MAILING DATE OF THE ADVISORY ACTION. IN NO EVENT WILL THE STATUTORY PERIOD FOR RESPONSE EXPIRE LATER THAN SIX MONTHS FROM THE DATE OF THIS FINAL ACTION.

Joseph L. Schofer

JOSEPH L. SCHOFER
SUPERVISORY PATENT EXAMINER
ART UNIT 155

dw
David Wu
July 26, 1994

MAIL ROOM
74 JAN 27 1995
PAT. & TRADEMARK OFF.

15X 2/28 061 150

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Pak-Wing Steve Chum, et al.

RECEIVED FEB 27 1995 GROUP 1500

Serial No.: 08/054,379 Group Art Unit: 1505

Filed: April 28, 1993 Examiner: D. Wu

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

"Express Mail" mailing label number TB 184 380 515
Date of Deposit January 27, 1995
I hereby certify that this paper or fee is being deposited with the United States Postal Service, with sufficient postage "Express Mail Post Office to Addressee" service under 37 CFR 1 10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D C. 20231
Jan Alverson
(Typed or printed name of person mailing paper or fee)
Jan Alverson
(Signature of person mailing paper or fee)

Hon. Commissioner of Patents & Trademarks
Washington, D.C. 20231

Sir:

EXTENSION OF TIME

Applicant(s) hereby request(s) that the shortened statutory period for response now set to expire October 27, 1994 in this application be extended for a period of three (3) month(s), the extended period then expiring on January 27, 1995. This is a second request for an extension of time.

Please charge $870.00 to our Deposit Account No. 04-1512. If this estimate is incorrect, please charge or credit our account accordingly. Two duplicate copies of this sheet are enclosed.

Respectfully submitted,

By L Wayne White

L. Wayne White
Registration No. 25,415
Address: Bldg. B-1211
Freeport, Texas 77541
Phone: 409-238-2149

LWW/jka
C-40,121-G
MM11291 02/13/95 08054379 04-1512 110 117 870.00CH

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office

Address : COMMISSIONER OF PATENTS AND TRADEMARKS
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED APPLICANT		ATTORNEY DOCKET NO.
08/054,379	04/28/93	CHUM	P	C40121G

15M2/0306

THE DOW CHEMICAL CO.
PATENT DEPARTMENT
BLDG. B-1211
DALLAS, TX 77541

WU, D

EXAMINER	
ART UNIT	PAPER NUMBER
1505	9

DATE MAILED: 03/06/95

NOTICE OF ABANDONMENT

This application is abandoned in view of:

1. ☒ Applicant's failure to respond to the Office letter, mailed 7/27/94 .
2. ☐ Applicant's letter of express abandonment which is in compliance with 37 C.F.R. 1.138.
3. ☐ Applicant's failure to timely file the response received _____ within the period set in the Office letter.
4. ☐ Applicant's failure to pay the required issue fee within the statutory period of 3 months from the mailing date of _____ of the Notice of Allowance.

   ☐ The issue fee was received on _____ .

   ☐ The issue fee has not been received in Allowed Files Branch as of _____ .

   In accordance with 35 U.S.C. 151, and under the provisions of 37 C.F.R. 1.316(b), applicant(s) may petition the Commissioner to accept the delayed payment of the issue fee if the delay in payment was unavoidable. The petition must be accompanied by the issue fee, unless it has been previously submitted, in the amount specified by 37 C.F.R. 1.17 (l), and a verified showing as to the causes of the delay.

   If applicant(s) never received the Notice of Allowance, a petition for a new Notice of Allowance and withdrawal of the holding of abandonment may be appropriate in view of Delgar Inc. v. Schuyler, 172 U.S.P.Q. 513.

5. ☐ Applicant's failure to timely correct the drawings and/or submit new or substitute formal drawings by _____ as required in the last Office action.

   ☐ The corrected and/or substitute drawings were received on _____ .

6. ☐ The reason(s) below.

David Wu

DAVID W. WU
PATENT EXAMINER
GROUP 1500

PTO-1432 (REV. 5-83)

PATENT APPLICATION SERIAL NO. 08/378998

U.S. DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE
FEE RECORD SHEET

SC13131 02/16/95 08378998 04-1512 130 101 730.00CH

PTO-1556
(5/87)

10

IN T NITED STATES PATENT AND TRADEMA FFICE 08/378998

Applicant(s): Pak-Wing Steve Chum, et al.
Serial No.: 08/054,379
Filed: April 28, 1993
Group Art Unit: 1505
Examiner: D. Wu
For: ~~FABRICATED ARTICLES MADE FROM ETHYLENE~~ POLYMER BLENDS

MAIL ROOM JAN 27 1995 PAT & TRADEMARK

BOX FWC
The Commissioner of Patents and Trademarks
Washington D.C. 20231

"Express Mail" mailing label number TB 184 380 515
Date of Deposit January 27, 1995
I hereby certify that this paper or fee is being deposited with the United States Postal Service, with sufficient postage "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231
Jan Alverson
(Typed or printed name of person mailing paper or fee)
Jan Alverson
(Signature of person mailing paper or fee)

Sir:

This is a request for filing a ☒ continuation ☐ divisional ☐ continuation-in-part) application under) 37 C.F.R. 1 62.

10/13

1. The following information is provided in accordance with 37 C.F.R. 1.62(h) (if different from originally filed or that stated in the Declaration and Power of Attorney presently of record) :
   (a) Title:
   (b) Names and addresses of Applicant(s):
2. Priority claim ☐ has ☒ has not been made pursuant to 35 USC 119, and if so, is identified as follows:
3. ☐ Cancel in this application claims _____ of the prior application before calculating the filing fee.
4. ☐ A Preliminary Amendment is enclosed.
5. ☒ The filing fee is calculated below.

For	Number Filed		Number Extra		Rate	Basic Fee $730.00
Total Claims	15	- 20 =	0	x	$ 22 00 =	0
Independent Claims	2	- 3 =	0	x	$ 76.00 =	0
Multiple Dependent Claims Presented					$240 00 =	0
Total Filing Fee ----------					=	$ 730

6. ☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment, to Account No. 04-1512. This sheet is enclosed in triplicate.

dw 3/16/94 B1

7. ☒ Amend the specification by inserting after Title of the Invention: -- Cross-Reference to Related Application -- This is a ☒ continuation ☐ divisional ☐ continuation-in-part of application Serial No. 08/054,379, filed April 28, 1993 (now abandoned)
8. ☐ A new declaration or oath is attached pursuant to 37 C F R 1.62(c)
9. ☐ New drawings are enclosed.
10. ☒ The inventorship of this application is the same as that of the prior copending U S application
    ☐ Fewer inventors are to be named in this application than in the prior copending U S application Delete _____ as inventor(s) in this application
    ☐ The inclusion of additional claimed subject matter in this application requires that additional inventor(s) be named The additional inventor(s) are identified in No. 1 above and a new oath or declaration is enclosed
11. ☐ Enter the amendment previously filed on _____, but unentered, in the prior application
12. ☐ A new Disclosure Statement is enclosed.
    ☒ Applicant(s)' Disclosure Statement is as filed with the parent application
13. ☒ The Power of Attorney in the prior application is to STEPHEN P KRUPP, Reg. No. 34,366, L Wayne White, Reg. No. 25,415; Glenn H. Korfhage, Reg. No. 27,204; Richard G. Waterman, Reg. No. 20,128
    Please address all communications to The Dow Chemical Company, Patent Department, B-1211, 2301 N. Brazosport Blvd., Freeport, Texas 77541
    a. ☒ The power appears in the original papers in the prior application
    b. ☐ The power does not appear in the original papers A copy of the power in the prior application is enclosed.
    c. ☒ Recognize as Associate Attorney Noreen D. Warrick whose Registration No. is 34,573 and whose phone number is 409-238-4510

1/27/95
(Date)

L Wayne White
L Wayne White
Registration No. 25,415
Phone 409-238-2149

C-40,121-AB
NDW/jka

UNITED STATES DEPARTMENT OF COMMERCE
Patent and Trademark Office
Address: COMMISSIONER OF PATENTS AND TRADEMARKS
Washington, D.C. 20231

SERIAL NUMBER	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.
08/378,998	01/27/95	CHUM	C-40,121-AB

EXAMINER: WU, D

ART UNIT	PAPER NUMBER
1505	11

15M2/0418

THE DOW CHEMICAL COMPANY
PATENT DEPARTMENT B 1211
2301 NORTH BRAZOSPORT BLVD
FREEPORT TX 77541

DATE MAILED: 04/18/95

This is a communication from the examiner in charge of your application.
COMMISSIONER OF PATENTS AND TRADEMARKS

☒ This application has been examined ☒ Responsive to communication filed on 1-27-95 ☒ This action is made final.

A shortened statutory period for response to this action is set to expire 3 month(s), 0 days from the date of this letter.
Failure to respond within the period for response will cause the application to become abandoned. 35 U.S.C. 133

Part I THE FOLLOWING ATTACHMENT(S) ARE PART OF THIS ACTION:

1. ☐ Notice of References Cited by Examiner, PTO-892.
2. ☒ Notice of Draftsman's Patent Drawing Review, PTO-948.
3. ☐ Notice of Art Cited by Applicant, PTO-1449.
4. ☐ Notice of Informal Patent Application, PTO-152.
5. ☐ Information on How to Effect Drawing Changes, PTO-1474..
6. ☐ _____.

Part II SUMMARY OF ACTION

1. ☒ Claims 1-8 and 17-23 are pending in the application.

   Of the above, claims _____ are withdrawn from consideration.

2. ☒ Claims 9-16 and 24-30 have been cancelled.

3. ☐ Claims _____ are allowed.

4. ☒ Claims 1-8 and 17-23 remain are rejected.

5. ☐ Claims _____ are objected to.

6. ☐ Claims _____ are subject to restriction or election requirement.

7. ☐ This application has been filed with informal drawings under 37 C.F.R. 1.85 which are acceptable for examination purposes.

8. ☐ Formal drawings are required in response to this Office action.

9. ☐ The corrected or substitute drawings have been received on _____. Under 37 C.F.R. 1.84 these drawings are ☐ acceptable; ☐ not acceptable (see explanation or Notice of Draftsman's Patent Drawing Review, PTO-948).

10. ☐ The proposed additional or substitute sheet(s) of drawings, filed on _____. has (have) been ☐ approved by the examiner; ☐ disapproved by the examiner (see explanation).

11. ☐ The proposed drawing correction, filed _____, has been ☐ approved; ☐ disapproved (see explanation).

12. ☐ Acknowledgement is made of the claim for priority under 35 U.S.C. 119. The certified copy has ☐ been received ☐ not been received ☐ been filed in parent application, serial no. _____ ; filed on _____.

13. ☐ Since this application apppears to be in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under Ex parte Quayle, 1935 C.D. 11; 453 O.G. 213.

14. ☐ Other

EXAMINER'S ACTION

PTOL-326 (Rev. 2/93)

15. The following is a quotation of the appropriate paragraphs of 35 U.S.C. § 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless --
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

16. The following is a quotation of 35 U.S.C. § 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.
>
> Subject matter developed by another person, which qualifies as prior art only under subsection (f) or (g) of section 102 of this title, shall not preclude patentability under this section where the subject matter and the claimed invention were, at the time the invention was made, owned by the same person or subject to an obligation of assignment to the same person.

17. Claims 1-8, and 17-23 are rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103 as obvious over WO '414.

18. In examples 1 and 2, and Table 1, WO '414 discloses polymer blends B1 and B2 made by ethylene polymers '006, and '013 wherein the MWD, melt index, and density fall into the same ranges as claimed. Although WO '414 fails to show the value of the slope of strain hardening coefficient of polymer '013, in view of other substantially similar physical properties, the examiner has a reasonable basis to believe that it inherently possesses said slope of strain hardening coefficient, and the polymer blends of WO '414 are not necessarily different from the claimed polymer blends. Since the examiner does not have proper equipment to carry out the analytical tests, the burden is on the applicants to prove the claimed polymer blends are necessarily different from those of WO '414 and unobvious thereof. ***In re Fitzgerald et al.*** 205 USPQ 594 (CCPA 1980).

19. Even if the properties of the polymer composition of the instant claims and the prior art examples are not the same, it would still have been obvious to one of ordinary skill in the art to make polymers having the claimed properties because it appears that the reference generically embrace the claimed polymer blends and the person of ordinary skill in the art would have expected all embodiments of the reference to work. See pages 6-7 of reference. Applicants have not demonstrated that the differences, if any, between the claimed product and the

products of the prior art examples give rise to unexpected results.

20. Applicant's arguments filed on May 18, 1994 have been fully considered but they are not deemed to be persuasive.

21. Applicants argue that part (A) of claimed polymer blends is a substantially linear ethylene/alpha-olefin interpolymer which is not a linear polymer but has "long chain branching", and part (B) is a heterogeneously branched ethylene polymer wherein less than 10 wt.% of a polymer fraction having SHC > about 1.3. However, the physical properties of '006 and '013 of WO '414 such as density, MWD, melt index are substantially similar to those set forth in the claims. Moreover, Applicants are reminded that the claims --not specification or examples-- define what Applicants regard as their invention. Here, the term "long chain branching" which applicants alleged as novelty of part (A) has never been set forth in the claims. Applicants also alleged that in Table 3, example 1 and 2 both have significantly higher dart impact and toughness than those of comparative example 3. However, none of the comparative experiments (including comparative example 3) are truly representative of the closest disclosure of WO '414.

The Chum's Declaration filed on May 18, 1994 has been considered but not deemed to be persuasive since there is no any comparative tests to show the unexpected results of the claimed polymer blends, mere conclusory statements are not entitled to probative weight. Since applicants have not met their burden to provide objective evidence demonstrating the claimed polymer blends are in fact differ from those of WO '414, the 102(b)/103 rejection is still deemed to be proper.

22. This is a Continuation of applicant's earlier application S.N. 08/054,379. All claims are drawn to the same invention claimed in the earlier application and could have been finally rejected on the grounds or art of record in the next Office action if they had been entered in the earlier application. Accordingly, **THIS ACTION IS MADE FINAL** even though it is a first action in this case. See M.P.E.P. § 706.07(b). Applicant is reminded of the extension of time policy as set forth in 37 C.F.R. § 1.136(a).

A SHORTENED STATUTORY PERIOD FOR RESPONSE TO THIS FINAL ACTION IS SET TO EXPIRE THREE MONTHS FROM THE DATE OF THIS ACTION. IN THE EVENT A FIRST RESPONSE IS FILED WITHIN TWO MONTHS OF THE MAILING DATE OF THIS FINAL ACTION AND THE ADVISORY ACTION IS NOT MAILED UNTIL AFTER THE END OF THE THREE-MONTH SHORTENED STATUTORY PERIOD, THEN THE SHORTENED STATUTORY PERIOD WILL EXPIRE ON THE DATE THE ADVISORY ACTION IS MAILED, AND ANY EXTENSION FEE PURSUANT TO 37 C.F.R. § 1.136(a) WILL BE CALCULATED FROM THE MAILING DATE OF THE ADVISORY ACTION. IN NO EVENT WILL THE

STATUTORY PERIOD FOR RESPONSE EXPIRE LATER THAN SIX MONTHS FROM THE DATE OF THIS FINAL ACTION.

23. Any inquiry concerning this communication or earlier communications from the examiner should be directed to David Wu whose telephone number is (703) 308-2450. The examiner can normally be reached on weekdays from 8:00 am to 4:00 pm.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Mr. Joseph Schofer, can be reached on (703) 308-2452. The fax phone number for this Group is (703) 305-5432.

Any inquiry of a general nature or relating to the status of this application or proceeding should be directed to the Group receptionist whose telephone number is (703) 308-2351.

David Wu
DAVID W. WU
PATENT EXAMINER
GROUP 1500

David Wu
April 14, 1995

PTO FORM 948
(REV. 7-92)

U.S. DEPARTMENT OF COMMERCE
Patent and Trademark Office

ATTACHMENT TO PAPER NUMBER	
APPLICATION NUMBER	054379

GROUP

NOTICE OF DRAFTSPERSON'S PATENT DRAWING REVIEW

THE PTO DRAFTSMEN REVIEW ALL ORIGINALLY FILED DRAWINGS REGARDLESS OF WHETHER THEY WERE DESIGNATED AS INFORMAL OR FORMAL. ADDITIONALLY, THE PATENT EXAMINER WILL ALSO REVIEW THE DRAWINGS FOR COMPLIANCE WITH THE REGULATIONS.

The drawings filed 4/28/93

A. ☐ are approved by the draftsperson.

B. ☒ are objected to by the draftsperson under 37 CFR 1.84 for the reason(s) checked below. The examiner will require submission of new, corrected drawings at the appropriate time. Corrected drawings must be submitted according to the instructions listed on the back of this Notice.

1. Paper and ink. 37 CFR 1.84(a)

☐ Sheet(s)_____Poor.

2. Size of Sheet and Margins. 37 CFR 1.84(b)

Acceptable Paper Sizes and Margins.

Margin	Paper Size: 8 1/2 by 14 inches	8 1/2 by 13 inches	DIN size A4 21 by 29.7 cm.
Top	2 inches	1 inch	2.5 cm.
Left	1/4 inch	1/4 inch	2.5 cm.
Right	1/4 inch	1/4 inch	1.5 cm.
Bottom	1/4 inch	1/4 inch	1.0 cm.

☒ Proper Size Paper Required. All Sheets Must be Same Size.
Sheet(s) 1-2

☐ Proper Margins Required.
Sheet(s) _____

☐ TOP ☐ RIGHT
☐ LEFT ☐ BOTTOM

3. Character of Lines. 37 CFR 1.84(c)

☐ Lines Pale or Rough and Blurred.
Fig(s)_____

☐ Solid Black Shading Not Allowed.
Fig(s)_____

4. ☐ Photographs Not Approved.

_____

☐ Comments;

5. Hatching and Shading. 37 CFR 1.84(d)

☐ Shade Lines are Required.
Fig(s) _____

☐ Criss-Cross Hatching Not Allowed.
Fig(s) _____

☐ Double Line Hatching Not Allowed.
Fig(s) _____

☐ Parts in Section Must be Hatched.
Fig(s) _____

6. Reference Characters. 37 CFR 1.84(f)

☐ Reference Characters Poor or Incorrectly Sized.
Fig(s) _____

☐ Reference Characters Placed Incorrectly.
Fig(s) _____

7. Views. 37 CFR 1.84(i) & (j)

☐ Figures Must be Numbered Properly.

☐ Figures Must Not be Connected.
Fig(s) _____

8. ☐ Identification of Drawings. 37 CFR 1.84(1)
Extraneous Matter or Copy Machine Marks Not Allowed. Fig(s) _____

9. ☐ Changes Not Completed from Prior PTO-948 dated _____

Telephone inquires concerning this review should be directed to the Chief Draftsperson at telephone number (703) 305-8404.

JF — Reviewing Draftsperson

8/28/93 — Date

Note: Any objection to the drawings made by the examiner will be communicated separately in an office action.

PTO Copy