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**WO 2005/017008 A1**

(54) Title: ALKYL ACRYLATE COPOLYMER MODIFIED ORIENTED POLYPROPYLENE FILMS, TAPES, FIBERS AND WOVEN AND NONWOVEN TEXTILES

(57) Abstract: Disclosed are films, tapes, slit film fibers and melt-spun fibers comprising (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and (b) from 1 to 30 weight % of at least one ethylene/ alkyl acrylate copolymer. The tapes can be stretched (i.e. uniaxially oriented) to provide monofilament slit film fibers. Also disclosed are processes for preparing such fibers. Also disclosed are nonwoven textiles prepared from the melt-spun fibers.

## TITLE OF THE INVENTION

ALKYL ACRYLATE COPOLYMER MODIFIED ORIENTED  
POLYPROPYLENE FILMS, TAPES, FIBERS AND WOVEN AND  
NONWOVEN TEXTILES

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## TECHNICAL FIELD

**[0001]** This invention relates to films, tapes and fibers comprising polypropylene modified with ethylene/alkyl acrylate copolymers. It also relates to woven and nonwoven textiles prepared from melt-spun fibers prepared from these modified polypropylene compositions. It also relates 10 to methods for preparing slit film fibers of these modified polypropylene compositions.

## BACKGROUND ART

**[0002]** Oriented films comprising polypropylene may be formed from the molten polymer by a number of methods known in the art. Films can 15 be oriented in one direction by hot-drawing in the machine direction with a tensioning device, and annealing. Films can also be oriented in two directions (machine direction and transverse direction) by suitable tensioning devices. Oriented polypropylene films are useful for a wide variety of packaging applications.

20 **[0003]** Fibers comprising polypropylene may be formed directly from the molten polymer by a number of methods known in the art, including melt-spinning, centrifugal spinning and melt-blowing.

**[0004]** Fibers also may be prepared from extruded polypropylene (PP) homopolymer or copolymer films. Flat films can be extruded into a cooling 25 water bath or onto chilled rolls for quenching. Alternatively, a tubular blown film can be extruded through an annular die and air-quenched. The quenched film is then knife-slit into tapes. The tapes are then stretched (i.e. uniaxially oriented) to several times their original length by hot-drawing in the machine direction with a tensioning device and annealing 30 the stretched tapes having controlled widths, for example, from about 2.5 mm to about 6.5 mm. The stretched tapes provide monofilament fibers.

**[0005]** Melt spun fibers can be incorporated into nonwoven textiles by many technologies, including dry laid, wet laid, air laid, spunbond,

spunlace, and needlepunch processes. The nonwovens so produced can be used in a wide range of applications including apparel, carpet backing, agrotextiles and geotextiles.

**[0006]** Polypropylene fibers can be used in twines or ropes or to 5 prepare yarns for carpets. Polypropylene fibers also can be woven or knitted into fabrics used in applications such as tarpaulins, liners, banners, sacking, carpet backing, agrotextiles and geotextiles.

**[0007]** To reduce the cost of the fabric woven from stretched slit-film tapes it is desirable to: 10

**[0008]** reduce the denier (similar width but lower thickness) of the tape;

**[0009]** increase the width of the tape (with similar thickness) and reduce the construction of the woven fabric by reducing the number of tapes per dimension (for example, from an 8 x 8 tapes/square inch to a 5 x 5 tapes/square inch construction); and/or

15 **[0010]** increase the amount of inert filler such as  $\text{CaCO}_3$  in the polypropylene formulation.

**[0011]** With conventional polypropylene compositions used in preparing tapes, it is rather difficult to achieve all the above objectives. Higher stretch ratios and reduced denier can result in unacceptable loss of 20 physical properties. Undesirable fibrillation of PP tapes under high stretch ratios must also be overcome.

**[0012]** Similarly, it is also desirable to provide oriented polypropylene films and fibers with improved mechanical properties such as tensile breaking load, tenacity (tensile breaking stress) and elongation to break.

25 **[0013]** Polypropylene fibers with improved mechanical properties have been prepared by adding small amounts of additives to polypropylene. For example, PCT Patent Application Publication WO 2003/048434 describes a method for making polypropylene monofilaments by adding 0.1 to 20 % of an additive consisting of a combination of lubricant, filler, 30 and heat stabilizer. European Patent EP0080274B1 describes methods for making polypropylene melt-spun fibers by adding 0.1 to 10 % of another polymer that is immiscible in a melt of the polypropylene (in particular, polyhexamethylene adipamide). Polypropylene has also been

modified by the addition of small amounts of liquid crystal polymers, polyethylene, polyethylene glycol and nylon 66 (*Journal of Applied Polymer Science*, 1986, 31(8), 2753-68.). Improvement of the mechanical properties of the modified compositions has been attributed to a lowering of spun orientation, or wind-up speed suppression.

5 **[0014]** Blends of polypropylene and ethylene/alkyl acrylate copolymers have been previously disclosed (see, for example, U.S. Patent Number 3,433,573).

#### DISCLOSURE OF THE INVENTION

10 **[0015]** The present invention relates to the enhancement of the mechanical properties such as tensile breaking load, tenacity (tensile breaking stress) and elongation to break (%) of stretched polypropylene films and tapes and enhancement of tenacity and elongation to break (%) of melt spun polypropylene fibers. These enhancements are achieved by 15 incorporating a small percentage (from 1 to 30, preferably from 1 to 15 weight %) of an ethylene/alkyl acrylate copolymer in the PP formulations used to prepare the films and fibers (the PP formulations may optionally contain other materials selected from fillers such as CaCO<sub>3</sub> and additives such as UV stabilizers, pigments etc.). Blending of ethylene/alkyl acrylate 20 copolymers with PP also improves the processability of the PP resin by, for example, increasing the melt strength of the molten PP resin. These blends provide improved stretchability and drawability for films and fibers over conventional polypropylene films and fibers. The drawability of tapes prepared from these blends is improved during manufacturing (i.e. the 25 blends allow increased draw ratios with no or reduced tape fibrillation). Benefits of the modified polypropylene blends include higher strength at the same tape or fiber denier with no loss in elongation at break.

**[0016]** The modified polypropylene blends can also provide cost reduction by allowing production of finer denier polypropylene fibers 30 having the same performance level as unmodified fibers at the same equipment throughput. Alternatively, higher throughput may be achieved at the same denier with improved performance. The blends described

herein will provide lower production downtime for most processes, resulting from fewer breaks.

**[0017]** An object of this invention is to improve the mechanical properties such as tensile breaking load, tenacity and elongation at break of polypropylene compositions used in films, melt-spun fibers, slit-film tapes and fibers there from by incorporating ethylene/alkyl acrylate copolymers into the PP resin formulations used for their preparation.

**[0018]** Another object of this invention is to improve or maintain the elastic properties of polypropylene homopolymer formulations loaded with higher levels of inert fillers such as  $\text{CaCO}_3$  by incorporating a certain level of ethylene/alkyl acrylate copolymers or using ethylene/alkyl acrylate copolymers as the resin vehicles (for filler loading) in the polypropylene formulations. The aim is to improve the stretchability of the films, tapes and slit film fibers without loss of tensile properties.

**[0019]** Another object of this invention is to improve the processability of PP formulations by incorporation of a small percentage of ethylene/alkyl acrylate copolymers. Generally, the cooling of a cast film (before slitting) by a chilled roll is less efficient than by using a chilled water bath. The stretchability of a film cooled on a chilled roll is poor, because the slower cooling results in a more crystalline film. The incorporation of ethylene/alkyl acrylate copolymers enhances the stretchability of the films and overcomes or reduces the fibrillation of the slit film tapes prepared from such films in subsequent drawing. It also enhances the drawability of melt spun fibers and enhances the tensile properties of the drawn fibers.

**[0020]** Another object of this invention is to enhance the tensile properties (tensile breaking load, tenacity and elongation at break) of the stretched tape with a lower denier.

**[0021]** Additional benefits of the modified polypropylene blends described herein include improved uniformity in thickness and width of films and fibers, reduced static, and improved abrasion resistance.

**[0022]** Accordingly, this invention provides a film prepared from a composition comprising

**[0023]** (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and

5 **[0024]** (b) from 1 to 30 weight % of at least one ethylene/ alkyl acrylate copolymer.

**[0025]** This invention also provides a slit film tape prepared from said film.

10 **[0026]** This invention also provides a fiber prepared by hot-drawing and annealing said slit film tape prepared from said film.

**[0027]** This invention further provides a melt-spun fiber prepared from a composition comprising

15 **[0028]** (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and

**[0029]** (b) from 1 to 15 weight % of at least one ethylene/alkyl acrylate copolymer.

20 **[0030]** This invention also provides nonwoven textiles prepared from melt-spun fibers as described above.

**[0031]** Benefits of using the modified polypropylene blends described herein include improvements in tensile strength, "low speed" perforation and "high speed" perforation.

25 **[0032]** This invention also provides films, slit film tapes, fibers (e.g. slit film fibers or melt-spun fibers) and nonwoven textiles wherein the composition described above further comprises (c) from 0.01 to 40 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.

30 **[0033]** This invention also provides processes for preparing slit film fibers comprising

**[0034]** (1) preparing a composition comprising

**[0035]** (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block

copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin;

**[0036]** (b) from 1 to 30 weight % of at least one ethylene/alkyl acrylate copolymer; and optionally

5 **[0037]** (c) from 0.01 to 40 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives;

**[0038]** (2) forming said composition into a film;

**[0039]** (3) slitting said film into tapes;

10 **[0040]** (4) hot-drawing and annealing a tape of step (3).

#### DETAILED DESCRIPTION OF THE INVENTION

**[0041]** "Copolymer" means polymers containing two or more different monomers. The terms "dipolymer" and "terpolymer" mean polymers containing only two and three different monomers respectively. The 15 phrase "copolymer of various monomers" means a copolymer whose units are derived from the various monomers.

**[0042]** As indicated above, the films, tapes and fibers of this invention are prepared from a composition comprising polypropylene resins modified with ethylene/alkyl acrylate copolymers.

20 **[0043]** Polypropylene (abbreviated PP) polymers include homopolymers, random copolymers, block copolymers and terpolymers of propylene. Copolymers of propylene include copolymers of propylene with other olefins such as ethylene, 1-butene, 2-butene and the various pentene isomers, etc. and preferably copolymers of propylene with

25 ethylene. Terpolymers of propylene include copolymers of propylene with ethylene and one other olefin. Random copolymers, also known as statistical copolymers, are polymers in which the propylene and the comonomer(s) are randomly distributed throughout the polymeric chain in ratios corresponding to the feed ratio of the propylene to the

30 comonomer(s). Block copolymers are made up of chain segments consisting of propylene homopolymer and of chain segments consisting of, for example, random copolymers of propylene and ethylene. The term "polypropylene" when used herein is used generically to refer to any or all of the polymers comprising propylene described above.

**[0044]** Polypropylene homopolymers or random copolymers can be manufactured by any known process. For example, polypropylene polymers can be prepared in the presence of Ziegler-Natta catalyst systems, based on organometallic compounds and on solids containing 5 titanium trichloride.

**[0045]** Block copolymers can be manufactured similarly, except that propylene is generally first polymerized by itself in a first stage and propylene and additional comonomers such as ethylene are then polymerized, in a second stage, in the presence of the polymer obtained 10 during the first. Each of these stages can be carried out, for example, in suspension in a hydrocarbon diluent, in suspension in liquid propylene, or else in gaseous phase, continuously or noncontinuously, in the same reactor or in separate reactors.

**[0046]** Additional information relating to block copolymers and to their 15 manufacture may be found particularly in chapters 4.4 and 4.7 of the work "Block Copolymers" edited by D. C. Allport and W. H. Janes, published by Applied Science Publishers Ltd in 1973.

**[0047]** The term "ethylene/alkyl acrylate copolymers" includes 20 copolymers of ethylene and alkyl acrylates wherein the alkyl moiety contains from one to six carbon atoms. Examples of alkyl acrylates include methyl acrylate, ethyl acrylate and butyl acrylate. "Ethylene/methyl acrylate (abbreviated EMA)" means a copolymer of ethylene (abbreviated E) and methyl acrylate (abbreviated MA). "Ethylene/ethyl acrylate (abbreviated EEA)" means a copolymer of ethylene (abbreviated E) and ethyl acrylate (abbreviated EA). "Ethylene/butyl acrylate (abbreviated EBA)" means a copolymer of ethylene (abbreviated E) and butylacrylate (abbreviated BA).

**[0048]** The relative amount of the alkyl acrylate comonomer 30 incorporated into ethylene/alkyl acrylate copolymer can, in principle, vary broadly from a few weight percent up to as high as 40 weight percent of the total copolymer or even higher. Similarly, the choice of the alkyl group can, again in principle, vary from a simple methyl group up to a six-carbon atom alkyl group with or without significant branching. The relative amount and choice of the alkyl group present in the alkyl acrylate ester

comonomer can be viewed as establishing how and to what degree the resulting ethylene copolymer is to be viewed as a polar polymeric constituent in the thermoplastic composition.

**[0049]** Preferably, the alkyl group in the alkyl acrylate comonomer has 5 from one to four carbon atoms and the alkyl acrylate comonomer has a concentration range of from 5 to 30 weight percent of the ethylene/alkyl acrylate copolymer, preferably from 10 to 25 weight %. Most preferably, the alkyl group in the alkyl acrylate comonomer is methyl.

**[0050]** Ethylene/ alkyl acrylate copolymers can be prepared by 10 processes well known in the polymer art using either autoclave or tubular reactors. The copolymerization can be run as a continuous process in an autoclave: ethylene, the alkyl acrylate, and optionally a solvent such as methanol (see U.S. Patent Number 5,028,674) are fed continuously into a stirred autoclave of the type disclosed in U.S. Patent Number 2,897,183, 15 together with an initiator. The rate of addition will depend on variables such as the polymerization temperature, pressure, alkyl acrylate monomer employed, and concentration of the monomer in the reaction mixture needed to achieve the target composition of the copolymer. In some cases, it may be desirable to use a telogen such as propane, to control the 20 molecular weight. The reaction mixture is continuously removed from the autoclave. After the reaction mixture leaves the reaction vessel, the copolymer is separated from the unreacted monomers and solvent (if solvent was used) by conventional means, e.g., vaporizing the nonpolymerized materials and solvent under reduced pressure and at an 25 elevated temperature.

**[0051]** Tubular reactor produced ethylene/alkyl acrylate copolymer can 30 be distinguished from the more conventional autoclave produced ethylene/alkyl acrylate as generally known in the art. Thus the term or phrase "tubular reactor produced" ethylene/alkyl acrylate copolymer, for purposes of this invention, denotes an ethylene copolymer produced at high pressure and elevated temperature in a tubular reactor or the like, wherein the inherent consequences of dissimilar reaction kinetics for the respective ethylene and alkyl acrylate comonomers is alleviated or partially compensated by the intentional introduction of the monomers

along the reaction flow path within the tubular reactor. As generally recognized in the art, such a tubular reactor copolymerization technique will produce a copolymer having a greater relative degree of heterogeneity along the polymer backbone (a more blocky distribution of comonomers),

5 will tend to reduce the presence of long chain branching and will produce a copolymer characterized by a higher melting point than one produced at the same comonomer ratio in a high pressure stirred autoclave reactor.

Tubular reactor produced ethylene/alkyl acrylate copolymers are generally stiffer and more elastic than autoclave produced ethylene/alkyl acrylate

10 copolymers.

**[0052]** Tubular reactor produced ethylene/alkyl acrylate copolymers of this nature are commercially available from E. I. du Pont de Nemours & Co., Wilmington, Delaware.

**[0053]** The actual manufacturing of the tubular reactor ethylene/alkyl acrylate copolymers as previously stated is preferably in a high pressure, tubular reactor at elevated temperature with additional introduction of reactant comonomer along the tube and not merely manufactured in a stirred high-temperature and high-pressure autoclave type reactor.

15 However, it should be appreciated that similar ethylene/alkyl acrylate copolymeric material can be produced in a series of autoclave reactors wherein comonomer replacement is achieved by multiple zone introduction of reactant comonomer as taught in U.S. Patent Numbers 3,350,372; 3,756,996; and 5,532,066, and as such these high melting point materials should be considered equivalent for purposes of this

20 invention.

**[0054]** To further illustrate and characterize the tubular reactor produced ethylene/alkyl acrylate copolymer relative to conventional autoclave produced copolymer, the following list of commercially available ethylene/methyl acrylate copolymers with associated melting point data

25 show that tubular EMA resins have considerably higher melting points versus autoclave EMA's due to a very different MA distribution along polymer chains:

**[0055]** Autoclave produced copolymers

**[0056]** EMA-A1 (21.5 wt% MA) mp = 76 °C

[0057] EMA-A2 (24 wt% MA) mp = 69 °C

[0058] EMA-A3 (20 wt% MA) mp = 80 °C

[0059] EMA-A4 (24wt% MA) mp = 73 °C.

[0060] Tubular reactor produced copolymers

5 [0061] EMA-T1 (25 wt% MA) mp = 88 °C

[0062] EMA-T2 (20 wt% MA) mp = 95 °C.

[0063] For additional discussion regarding the differences between tubular reactor produced and autoclave produced ethylene/alkyl acrylate copolymers, see Richard T. Chou, Mimi Y. Keating and Lester J. Hughes, 10 "High Flexibility EMA made from High Pressure Tubular Process", Annual Technical Conference - Society of Plastics Engineers (2002), 60th(Vol. 2), 1832-1836. CODEN: ACPED4 ISSN:0272-5223; AN 2002:572809; CAPLUS.

[0064] Ethylene/alkyl acrylate copolymers suitable for use in this 15 invention are available from DuPont. See Table A for specific examples of tubular reactor produced ethylene/alkyl acrylate copolymers available from DuPont.

Table A

Ethylene Alkyl Acrylate Copolymer	Alkyl acrylate wt%	Melt Flow (g/10min)
EMA	25	0.4
EMA	25	0.6
EMA	9	2
EMA	14	2
EMA	18	2
EMA	24	2
EMA	9	6
EMA	20	8
EMA	13	9
EEA	12	1
EEA	16	1
EEA	15	6
EEA	15	7
EBA	7	1.1
EBA	7	1.1
EBA	17	1.5
EBA	17	1.8
EBA	27	4
EBA	17	7

**[0065]** The ethylene/alkyl acrylate copolymers useful in the present invention can vary significantly in molecular weight as witnessed by EMA having a melt index numerically in terms of a fraction up to about ten. The specific selection of the grade of ethylene/alkyl acrylate copolymer component(s) to be used to modify polypropylene will be influenced by balancing factors such as melt indices of the modifier and the polypropylene, draw temperature related to the respective softening points of the ethylene/alkyl acrylate copolymer and the polypropylene, and the draw profile (draw rate and draw ratio) contemplated. Other factors to be considered in the selection of the ethylene/alkyl acrylate copolymer

include increased elastic recovery associated with higher relative molecular weight copolymer (such as an E/25 weight % MA with a 0.7 MI) and the pragmatic ability to more easily blend with fillers (see below) with a relatively lower molecular copolymer (such as an E/20 weight % MA with an 8 MI).

5 **[0066]** Compositions of ethylene/alkyl acrylate copolymers and polypropylene useful in this invention may be prepared by dry blending, pellet blending, melt blending, by extruding a mixture of the various constituents, and other mixing processes known in the art.

10 **[0067]** The compositions useful in this invention may optionally further comprise fillers such as calcium carbonate (CaCO<sub>3</sub>), in quantities that may be up to 30 to 40 weight %, of a tape composition. For example, from 0.01 to 20 weight %, from 0.1 to 15 weight %, from about 2 to about

15 10 weight % CaCO<sub>3</sub> may be present in some slit film tapes. The ethylene/alkyl acrylate copolymers as described herein provide enhanced compatibility between the polypropylene base resin and optional filler. The enhanced compatibility may provide reduced tendency of "dusting" caused by separation of the filler and the polypropylene resin, lower CaCO<sub>3</sub>

20 particulates in the air during processing operations, and reduced wear on weaving or knitting equipment. Use of ethylene/alkyl acrylate copolymer modifiers as described herein may also provide for higher filler loading.

25 **[0068]** The compositions useful in this invention may optionally further comprise other additives such as delustrants such as TiO<sub>2</sub>, UV stabilizers, pigments, etc. These additives are well known in the art of films, slit film fibers and melt spun fibers. These conventional ingredients may be present in the compositions according to this invention, from 0.01 to 20 weight %, preferably from 0.1 to 15 weight %.

30 **[0069]** The optional incorporation of such conventional ingredients into the compositions comprising polypropylene modified with ethylene/alkyl acrylate copolymer can be carried out by any known process. This incorporation can be carried out, for example, by dry blending, by extruding a mixture of the various constituents, by the conventional masterbatch technique, or the like. A typical masterbatch may comprise

from 75 to 90 weight % of CaCO<sub>3</sub>. Of note is the use of a masterbatch comprising CaCO<sub>3</sub> and the ethylene/alkyl acrylate copolymer modifier.

**[0070]** The thermoplastic compositions described herein are suitable for preparation of films and fibers by extrusion processing.

5   **[0071]** Films of this invention, from which slit film tapes and fibers may be formed, can be made by virtually any method of film forming known to those skilled in this art. As such, the film and film structures can be typically cast, extruded, co-extruded and the like including orientation (either uniaxially or biaxially) by various methodologies (e.g., blown film, 10 mechanical stretching or the like). It should be appreciated that various additives as generally practiced in the art can be present in the respective film layers including the presence of tie layers and the like, provided their presence does not substantially alter the properties of the film or film structure. Thus, it is contemplated that various additives such as 15 antioxidants and thermal stabilizers, ultraviolet (UV) light stabilizers, pigments and dyes, fillers, delustrants, anti-slip agents, plasticizers, other processing aids, and the like may be advantageously employed.

**[0072]** In one embodiment, the film is formed by an extrusion process that causes the polymer chains in the film to be generally aligned in the 20 direction of extrusion. Linear polymers, after being highly oriented uniaxially possess considerable strength in the orientation direction, but less strength in the transverse direction. This alignment can add strength to the film in the direction of extrusion, which corresponds to the length dimension of the slit film yarns. Alternatively, the film may be formed 25 through a blowing process known to those skilled in this art.

**[0073]** This invention provides a film prepared from a composition comprising polypropylene and ethylene/alkyl acrylate copolymers. The film can be unoriented, oriented in a uniaxial direction (e.g. machine direction), or oriented in a biaxial direction (e.g. machine direction and 30 transverse direction).

**[0074]** The films are useful in a wide variety of packaging applications including shrink film. As indicated above, the films are also useful for preparing slit film tapes and fibers.

**[0075]** Preferred films of this invention include:

**[0076]** Preferred 1. The film wherein said alkyl acrylate is present in said ethylene/alkyl acrylate copolymer in a range from about 5 to about 30 weight %.

5 **[0077]** Preferred 2. The film of Preferred 1 wherein said alkyl acrylate is present in said ethylene/ alkyl acrylate copolymer in a range from about 10 to about 25 weight %.

**[0078]** Preferred 3. The film of Preferred 2 wherein said alkyl acrylate is selected from the group consisting of methyl acrylate, ethyl 10 acrylate and butyl acrylate.

**[0079]** Preferred 4. The film of Preferred 3 wherein said alkyl acrylate is methyl acrylate.

15 **[0080]** Preferred 5. The film of any of Preferred 1 through Preferred 4 wherein component (b) is present in an amount from 2 to 30 weight %, alternatively from 2 to 20 weight %, alternatively from 2 to 10 weight % of the total composition.

**[0081]** Preferred 6. The film of any of Preferred 1 through Preferred 5 further comprising

20 **[0082]** (c) from 0.01 to 40 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.

**[0083]** Preferred 7. The film of Preferred 6 wherein component (c) is present in an amount between 0.1 and 15 weight %.

25 **[0084]** Preferred 8. The film of any of Preferred 1 through Preferred 7 that is prepared by extrusion of said composition into a cooling water bath for quenching.

**[0085]** Preferred 9. The film of any of Preferred 1 through Preferred 7 that is prepared by extrusion of said composition onto chilled rolls for quenching.

30 **[0086]** Preferred 10. The film of any of Preferred 1 through Preferred 7 that is prepared by extrusion of said composition through an annular die into a tubular blown film that is air-quenched.

**[0087]** Preferred 11. The film of any of Preferred 1 through Preferred 10 wherein said composition comprises a tubular reactor produced ethylene/ alkyl acrylate copolymer.

5 **[0088]** This invention also provides a slit film tape prepared from films of this invention. Preferred tapes are those prepared from the preferred films above.

**[0089]** This invention also provides a fiber prepared by hot-drawing and annealing said slit film tape prepared from said film. Preferred fibers are those prepared from tapes prepared from the preferred films above.

10 **[0090]** This invention also provides processes for preparing slit film fibers. Preferred processes are those utilizing the preferred films above.

**[0091]** As indicated, the slit film fibers are formed of a polymeric material that is formed into a film, slit into tapes and hot-drawn.

15 **[0092]** Woven or knitted textiles can be prepared from slit film fibers as described above. Preferred woven or knitted textiles are prepared from the preferred fibers above.

**[0093]** Processes to manufacture slit film fibers based on polypropylene polymers by slitting films extruded from compositions comprising such polymers are well known. Common usages for which 20 these slit film monofilament fibers are intended include rope and string manufacture, fibers for synthetic lawns, woven fabric for sacking and, in particular, heavy-duty sack construction, primary and secondary carpet backing, geotextiles, agrotextiles, industrial fabric, strapping, belting, apparel, diapers, netting, woven containers, and the like.

25 **[0094]** The manufacture of the slit film fiber itself from the compositions described above can be carried out according to any known methods. It is possible, for example, to manufacture a primary film by extruding the said compositions using so-called "blown film" or "flat die" methods. A blown film is prepared by extruding the polymeric composition through an 30 annular die and expanding the resulting tubular film with an air current to provide a blown film. Cast flat films are prepared by extruding the composition through a flat die. The film leaving the die is cooled by at least one roll containing internally circulating fluid (a chill roll) or by a water bath to provide a cast film. A film of this invention would have a width, for

example, of about 60 cm (two feet). This primary film is then slit into slit film tapes that are drawn into monofilament fibers before being reeled up.

**[0095]** The slit film tapes can be produced with a slitting apparatus which comprises a support frame; a plurality of substantially planar cutting blades, each of which includes opposed cutting edges and opposed ends; a mounting structure for mounting the cutting blades to the support frame; and a feed roll attached to the support frame and configured to feed film in a downstream direction over the exposed cutting edges of the blades.

5 The mounting structure is configured to mount the cutting blades in substantially aligned, parallel and spaced apart relationship, wherein the blades are mounted such that each blade has one of its cutting edges exposed for cutting, and wherein the cutting edges of adjacent blades are spaced apart from each other between about 6 mm and 8 mm.

**[0096]** After slitting the film into tapes as described above, the drawing 10 operation would be done over a span of from 3 to 6 meters (ten to twenty feet) in an oven heated to a temperature effective to soften the film so as to facilitate the drawing operation. What typically takes place is that the film, at the beginning of the path through the oven is cold and is progressively heated and softened as it passes through the oven.

15 Necking takes place at a neckline that is at a certain distance from the entrance to the oven. The location of the necking zone depends on a number of factors including the rate of stretching, the temperature of the oven, and the nature and thickness of the film material. A typical pre-stretched tape has a thickness about 120 microns and a width of about 6

20 mm to 8 mm. After stretching, the final tape has a thickness of about 30 to 50 microns and a width of about 2.5 mm to 3 mm. Tapes can be made wider or narrower for certain purposes. For example, fibers for reduced end count woven fabric can have a final width of from about 4 mm to about 6 mm; polypropylene strapping can have a final width of about 10 mm to

25 30 15 mm.

**[0097]** The drawing ratio will generally be in the range of from about 2:1 to about 16:1 and a typical drawing ratio for some polymers would be from about 4:1 to about 10:1. The drawing ratio for woven tape is preferably from about 5:1 to about 8:1. The drawing ratio for wider strapping tape is

typically from about 10:1 to about 15:1. The distance over which longitudinal drawing takes place will vary with the technique used. In the short-draw the stretching takes place over a distance of a few inches, other techniques involve much greater distances. After hot-drawing, the 5 resulting monofilament fibers for woven tape would typically have a denier of from about 700 to about 1700. Polypropylene strapping would have a denier of from about 3000 to about 6000.

10 [0098] The mechanical properties such as tenacity, tensile breaking load, elongation at break and denier of the tapes of this invention can be balanced by adjusting various parameters including

[0099] • resin formulation design (base resin, level and types of additives such as  $\text{CaCO}_3$ , UV stabilizers, pigment added);

[00100] • amount and type of ethylene/ alkyl acrylate used;

15 [00101] • film and tape processing equipment (quenching, slitting, drawing and annealing configuration); and

[00102] • processing conditions (extruder screw configuration, temperature profile and polymer throughput, stretch and annealing temperatures and profiles, line speed etc).

20 [00103] Typically, a manufacturing facility for preparing slit film yarns will have a limited ability to modify equipment and processing conditions.

Therefore, the ethylene/alkyl acrylate modification of the polypropylene resin of this invention can provide significant improvement in mechanical properties of the films, tapes and yarns prepared there from.

25 [00104] As noted above, fibers may also be prepared directly from extrusion processes including centrifugal spinning, melt-spinning, spunbonding and melt-blowing.

[00105] In centrifugal spinning, fibers are formed as a polymer melt is accelerated from a rapidly rotating source. Molten material from a furnace is transferred into a rotating spinner, and the fibers are produced as 30 centrifugal force extrudes the material through small holes in the side of the spinning device.

[00106] In melt spinning, the fiber-forming substance is melted for extrusion through the spinneret and then directly solidified by cooling.

Melt spun fibers can be extruded from the spinneret in different cross-sectional shapes (round, trilobal, pentalobal, octalobal, and others). In-line drawing is effected by wrapping the moving thread line around sets of rotating rolls running at controlled temperature and speeds. Depending on

5 the specific melt spinning process and subsequent processing steps, product can be collected as monofilaments, yarn, tow or nonwoven (e.g. spunbond). See Fibers from Synthetic Polymers, Rowland Hill, ed., Elsevier Publishing Co., NY, 1953 for a general reference regarding melt spun fibers.

10 **[00107]** Spunbonding is the direct laydown of nonwoven webs from fibers as they are melt-spun. Continuous filaments are extruded through a spinneret, accelerated (via rolls or jets) and laid down onto a moving belt to form a nonwoven sheet. Bonding occurs at molten fiber crossover points.

15 **[00108]** Meltblowing is another direct laydown process in which fibers are extruded through a die tip, attenuated (and fractured) by hot, high velocity air, and deposited onto a moving belt or screen to form a web of fine (low denier) fibers.

**[00109]** Both spunbond (S) and meltblown (M) webs, after being formed, can be further bonded and/or patterned by calendering. Multiple layered nonwovens (e.g. SMS, SMMS, SMMMS) can also be prepared from fibers of this invention.

**[00110]** This invention further provides a melt-spun fiber prepared from a composition as described above. Preferred melt-spun fibers include:

20 **[00111]** Preferred A. The fiber wherein said alkyl acrylate is present in said ethylene/alkyl acrylate copolymer in a range from about 5 to about 30 weight %.

**[00112]** Preferred B. The fiber of Preferred A wherein said alkyl acrylate is present in said ethylene/alkyl acrylate copolymer in a range from about 10 to about 25 weight %.

30 **[00113]** Preferred C. The fiber of Preferred B wherein said alkyl acrylate is selected from the group consisting of methyl acrylate, ethyl acrylate and butyl acrylate.

**[00114]** Preferred D. The fiber of Preferred C wherein said alkyl acrylate is methyl acrylate.

5 **[00115]** Preferred E. The fiber of any of Preferred A through Preferred D wherein component (b) is present in an amount from 2 to 10 weight %.

**[00116]** Preferred F. The fiber of any of Preferred A through Preferred E further comprising

10 **[00117]** (c) from 0.01 to 15 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.

**[00118]** Preferred G. The fiber of Preferred F wherein component (c) is present in an amount between 0.1 and 5 weight %.

15 **[00119]** Preferred H. The fiber of any of Preferred A through Preferred G wherein said composition comprises a tubular reactor produced ethylene/ alkyl acrylate copolymer.

**[00120]** Nonwoven textiles can be prepared from melt-spun fibers as described above. Preferred nonwoven textiles are prepared from the preferred fibers above.

20 **[00121]** Woven or knitted textiles can be prepared from melt-spun fibers as described above. Preferred woven or knitted textiles are prepared from the preferred fibers above.

**[00122]** Oriented films are used in a wide variety of packaging applications. As described above, films of this invention can be also used to prepare slit tape fibers.

25 **[00123]** Fibers prepared as described herein, including slit tape fibers are useful for preparing cords, twines or ropes. A number of fibers are joined together by, for example, twisting, braiding, interlacing and the like to form a cord. Twines, in general, contain a lower number of fibers and are smaller in diameter than ropes. These cords, twines or ropes may be

30 roughly circular or flattened in cross section. Cords and twines can be used for shoelaces, straps for bags, briefcases and the like, and can be used in packaging applications. Ropes can be used in a wide variety of industrial and marine applications. Cords, twines and ropes may also be

further interlaced (such as by knitting) to prepare nets having a relatively open structure, such as fishing nets, cargo nets and the like.

**[00124]** Slit tape fibers of this invention can be used as monofilament fibers for carpet tufting, synthetic lawns, matting and the like. They can 5 also be used as strapping.

**[00125]** Woven or knitted textiles can be prepared from the slit tape fibers or melt-spun fibers as described above. In general, woven fabrics may have a tighter construction than knitted fabrics. Polypropylene yarns prepared as described herein can be woven into fabrics used in 10 applications such as filters, tarpaulins, awnings, canopies, banners, construction (e.g. roofing) membranes, machine belts, liners for luggage or packaging, heavy duty sacking, carpet backing, upholstery, apparel, agrotextiles (for use in seed control, weed control, gardening, greenhouses and silage) and geotextiles (for erosion control and soil 15 conservation). Knitted fabrics can be used for sacking for use in carrying bulky materials such as groceries, firewood and the like; and construction, industrial and fishing netting.

**[00126]** Woven fabrics using the modified polypropylene compositions as described herein have a lower slip tendency, particularly for heavy duty 20 sacks.

**[00127]** Nonwoven fabrics of this invention can be used in diapers and other items used for personal hygiene such as adult incontinence and feminine hygiene products, medical apparel such as hats, gowns, booties, personal protective equipment including masks and the like, hygiene 25 protective furnishings such as drapes, covers, blankets and the like, packaging, durable paper, wipes, wraps, banners, carpet backing, filtering, geotextiles, agrotextiles, upholstery, apparel, filters, liners, or construction wrap for heat and moisture control in buildings.

**[00128]** For carpet backings, the shrink of the textile is important (for 30 example, shrinkage less than 2.5% at 45 °C. is desirable). Woven and nonwoven carpet backings are typically used as a primary backing for carpets to provide strength, dimensional stability and form to the carpet. They can be prepared from slit tape fibers or spunbonded fabrics of this invention.

**[00129]** Secondary carpet backings can be used to provide a substrate to which the carpet yarns are secured. They can be prepared from nonwoven materials. Melt spun fibers of this invention are useful for preparing nonwoven substrates useful as secondary carpet backing.

5 **[00130]** Geotextiles can be used in roads under the gravel and paving layers to improve road quality. Geotextile fabrics are typically manufactured by weaving PP tapes having approximately 2.5 mm width. Geotextiles are also prepared from spun bonded nonwoven material derived from melt-spun fibers. Shrinkage requirements for geotextiles are

10 10 not as stringent as those for carpet backings. However, resistance to perforation is extremely important for geotextiles.

**[00131]** While the invention has been particularly shown and described with reference to the preferred embodiments thereof, it will be understood by those skilled in the art that various changes in form and details may be

15 15 made without departing from the spirit and scope of the invention.

**[00132]** The following Examples are merely illustrative, and are not to be construed as limiting the scope of the invention described and/or claimed herein.

#### EXAMPLES

20 **[00133]** Slit Film Tapes

**[00134]** Trial 1

**[00135]** The trial was conducted on an extrusion casting line using the resin formulations indicated below. The film was cast onto a chilled (quench) roll. It was then slit into tapes and subsequently stretched 6 or

25 25 7X into 5 mm tapes in an annealing oven. The stretching and annealing of tapes were carried out simultaneously. Samples were taken and tested for tensile strength after the manufacturing procedure had been running for 15 minutes. A 250 mm length of 5.0 mm wide tape was stretched on an Universal Tensile Test Machine with a rate of cross head separation of

30 30 200 mm/min. The results are summarized in Table 2.

**[00136]** Materials Used

**[00137]** PP-1: Polypropylene homopolymer with a melt index (MI) of 3 g/10min (ASTM D-1238, 230 °C using a 2.16 Kg mass) available from Titan Petrochemical (M) Sdn Bhd.

5 **[00138]** EMA-1: Ethylene/alkyl acrylate copolymer with 24 weight % methyl acrylate with MI of 2 g/10min (ASTM D-1238, 190 °C using a 2.16 Kg mass).

**[00139]** Calcium Carbonate as filler.

**[00140]** Resin formulations:

10 **[00141]** Comparative Example C1: PP-1 (99%) + CaCO<sub>3</sub> (1%)

**[00142]** Example 2: PP-1 (89%) + CaCO<sub>3</sub> (1%) + EMA-1 (10%)

**[00143]** Example 3: PP-1 (94%) + CaCO<sub>3</sub> (1%) + EMA-1 (5%)

Table 2

	Comparative	Example C1	Example 2	Example 2	Example 3
Stretch ratio	6	6.12	7.05	6	
Denier	--	963	1178	1058	
Tensile Breaking	--				
Load (Kgf)		5.02	6.05	5.24	
Tenacity (g/denier)	--	5.212	5.136	4.953	
Elongation at break	--	31	27	32	
	(%)				

15 **[00144]** The run using Comparative Example C1 (0% EMA-1) was discontinued since processability was poor and unstable under the conditions used for Examples 2 and 3 (10% and 5% EMA-1). Fibrillation of tapes prepared from Comparative Example 1 was encountered during the trial even at a stretch ratio of 6.

20 **[00145]** Under normal production conditions with pure PP formulation (see Comparative Example 1), the cast film is unstable at high extruder throughput (for example, extruder screw speeds above 180 rpm). The

machine throughput is low and hence the line productivity is also reduced. Furthermore, if draw ratio is increased beyond about 6, fibrillation of tapes starts to occur and the width of the tapes is very inconsistent.

5 [00146] By incorporating 5 to 10% of EMA resin (for example EMA-1) into the PP formulation, the production can be run very smoothly at high throughput and the cast film from the extruder is very stable. The new formulation has better processability and improved melt strength.

10 [00147] The slit tapes can be stretched to 6 and 7 times with no tendency to fibrillate. The mechanical properties of the oriented tapes such as tenacity, tensile breaking load and elongation at break are satisfactory.

[00148] Trial 2

15 [00149] The trial was conducted on an extrusion casting line. The film quenched in a chilled water bath. It was then slit into tapes and subsequently stretched at least 6X into 3 mm tapes in an annealing oven. The stretching and annealing of tapes were carried out simultaneously. Samples were taken and tested for tensile strength after the manufacturing procedure had been running for 15 minutes. A 250 mm length of 3.0 mm wide tape was stretched on a Universal Tensile Test 20 Machine with a rate of cross head separation of 200 mm/min. The results are summarized in Table 3.

[00150] Materials Used

25 [00151] PP-2: polypropylene homopolymer resin with a melt index (MI) of 3 g/10min(ASTM D-1238, 230°C / 2.16Kg), available as PD855 from Titan Petrochemical (M) Sdn Bhd.

[00152] Ethylene/alkyl acrylate copolymer with 24 weight % methyl acrylate (EMA-1) with MI of 2 g/10min (ASTM D-1238, 190°C / 2.16Kg).

[00153] White master batch, a colorant/filler comprising CaCO<sub>3</sub>.

[00154] Resin formulations

30 [00155] Comparative Examples C4 and C6: PP-2 (95%) + white master batch (5%)

**[00156]** Examples 5 and 7 through 9: PP-2 (90%) + white master batch (5%) + EMA-1 (5%)

Table 3

	Compar- ative Example	Example 5	Compar- ative Example	Example 7	Example 8	Example 9
	C4		C6			
Stretch ratio	6	6	6	6	8	7
Denier	886	858	1075	1096	1016	736
Tensile						
Breaking	4.8	5.1	5.6	5.8	6.7	4.6
Load (Kgf)						
Tenacity (g/denier)	5.4	6.0	5.2	5.3	6.8	6.2
Elongation at break (%)	31	33	30	41	26	28

5 **[00157]** These trial results confirmed that the addition of 5% of ethylene/alkyl acrylate copolymer in the PP has improved the mechanical properties such as tensile breaking load, elongation at break and tenacity of oriented PP tapes.

10 **[00158]** Comparing tapes with similar average deniers (Comparative Example C4 and Example 5, Comparative Example C6 and Example 7), tapes produced from PP with 5% EMA incorporated have shown an improvement in the average tensile breaking load, average elongation at break and average tenacity than standard PP tapes stretched under a similar stretch ratio.

15 **[00159]** The improvement in the average tensile breaking and average tenacity of the modified PP tapes (Example 8) under a higher stretch ratio (8X) was even more significant as compared to standard (Comparative Example C6) or modified PP tapes (Example 7) produced at the standard stretch ratio (6X).

20 **[00160]** Modified PP tapes with a lower average denier (Example 5) have tensile properties nearly as high as standard PP tapes with a higher average denier (Comparative Example C6). Modified PP low denier tapes

(Example 9) prepared with high stretch ratio (7X) have similar average tensile breaking load and elongation at break but much higher tenacity than the standard PP tapes (Comparative Example C4).

**[00161] Trial 3**

5   **[00162]** In these trials the fibers were made in a manner suitable for incorporation into woven fabric suitable for geotextiles.

**[00163]** PP-3: A base polypropylene material is obtained by blending polypropylene (Amoco GA 02, MFI: 1.8) with 2 weight percent of a color additive (Ampacet Noir 190826) and 3.5 weight percent of a  $\text{CaCO}_3$  filler 10 (Multibase ME 50004), designated Comparative Example C10. The base material PP-3 is further blended with 3 weight percent of a polyolefin copolymer (Booster PO) to prepare Comparative Example C11. The base material PP-3 is blended with 5 weight percent or 10 weight percent of EMA-1 to prepare Examples 12 and 13.

15   **[00164] Processing conditions**

**[00165]** Extrusion temperature:                   255 °C

**[00166]** Oven (stretching):                       163 °C

**[00167]** Chill roll:                                   27 °C

**[00168]** Input:                                       270-300 kg/hr

20   **[00169]** Cast film width:                       1250 mm

**[00170]** Thickness of the cast film:               175-180  $\mu\text{m}$

**[00171]** Tape width before stretch:               6 mm

**[00172]** Tape width after stretch:               approximately 2.5 mm

**[00173]** Processing started with 5% EMA-1 and a stretch ratio of 7. The 25 stretch ratio was then increased to 7.4 and 7.8 to improve the strength.

Table 4

	Stretch Ratio	TEX [g/km]	Strength [cN]/TEX	Elongation
C10	6	172	42	30
C11	7	144	47-49	29
12	7	140	45	29.5
12	7.4	142	46	30.4
12	7.8	142	49.5	30
12	7.8	142	48	29.5
12	7.8	148	49	30.8
12	7.8	142 <sup>1</sup>	48.8	28.8
12	7.8	143 <sup>1</sup>	46.3	27.4
13	7.8	143	48.4	27.5
13	7.8	143.6	46.2	28.1
13	7.8	142.4	48	28.5
13	7.8	143.8	47	27.3
13	7.8	143.2	49	28.4
13	7.8	140	49	28
13	8	142.8	50.7	32.5
13	8	141.4	47.8	30.2
13	8.2	138	53.5	34.8
13	8.2	138.6	49.5	31.7

Notes: 1. After 2 hours. 2. Adjustment of stretch rolls. 3. Oven temperature of 160 °C.

**[00174]** Compared to polypropylene (Example C10), tapes prepared from a composition of this invention demonstrated a higher strength and similar elongation at a higher stretch ratio. Compared to polypropylene plus 3 weight % Booster PO (Example C11), tapes prepared from a composition of this invention demonstrated slightly higher elongation and strength at a higher stretch ratio.

10 **[00175]** In addition, using an ethylene/alkyl acrylate modified composition as described herein allowed for slightly lower energy consumption and slightly lower pressure at the extruder.

**[00176]** By incorporating ethylene/alkyl acrylate copolymers in PP formulations, one can produce

**[00177]** • Highly drawn, low denier tapes with very high tensile breaking load and tenacity without significant loss of elongation; or

5 **[00178]** • Highly drawn, high denier (wide but thin) tapes with good mechanical properties for cost saving in woven fiber bag or lamination substrate construction.

**[00179]** Spun Fibers

**[00180]** Trial 4

10 **[00181]** The purpose of this trial was to compare the spinning performance (maximum drawability) and physical properties of PP yarns vs. PP/EMA blend yarns. Yarns were drawn on the spinning machine between a pair of heated rolls, wound up, and tested. The PP/EMA blends were melt blended prior to spinning.

15 **[00182]** Materials Used

**[00183]** Comparative Example C14: 100% PP-4 (Profax® 6323)

**[00184]** Example 15: 95%/5% blend of PP-4 with EMA-1

**[00185]** Example 16: 90%/10% blend of PP-4 with EMA-1

**[00186]** Spinning conditions

20 **[00187]** Block temperature 245 °C

**[00188]** Spinneret 10 holes, 0.015 inch x 0.060 inch

**[00189]** Feed Roll Temperature 60 °C

**[00190]** Feed Roll Speed 500 yards per minute (ypm)

**[00191]** Draw Roll Temp 80 °C

25 **[00192]** Draw Roll Speed was variable to determine maximum draw (for example, 1500 ypm was used to obtain a 3X draw).

Table 5

Ex.	Draw Ratio	Yarn Denier	Tenacity (gpd)		Elongation (%)		Modulus (gpd)	
			Ave	SD	Ave.	SD	Ave.	SD
C14	3.0	89.8	3.23	0.11	103.38	9.78	34.4	2.5
C14	3.8	72.3	4.15	0.24	66.65	6.59	34.3	2.2
15	3.0	90.0	3.72	0.16	116.51	6.36	26.3	2.4
15	4.0	67.7	4.64	0.27	60.82	7.23	41.4	4.0
16	3.0	89.8	3.29	0.24	119.03	10.81	21.1	3.2
16	4.0	68.4	4.47	0.46	63.14	5.84	38.7	7.1

[00193] Addition of an ethylene/alkyl acrylate copolymer increases the residual elongation and decreases initial modulus in single-stage-drawn melt spun polypropylene fibers (indicating better drawability). Addition also increases the maximum spinning draw ratio leading to desirably increased strength and/or decreased denier.

**[00194] Trials 5 and 6**

[00195] Two full-day spinning trials were carried out. The first trial was done using blends made on a twin-screw mixing unit (melt blends). The second trial was done using blends of the polymers mixed in a tumble blender (dry blends). Blends with 5, 10 and 15 weight % of EMA-1 were prepared.

[00196] Melt blends prepared on twin-screw extruder and run during the first trial:

[00197] Comparative Example C17: 100% PP-5 (polypropylene with MFI of 4).

[00198] Example 18: 95%/5% blend of PP-5 with EMA-1.

[00199] Example 19: 90%/10% blend of PP-5 with EMA-1.

[00200] Dry blends prepared with a tumble blender and run during the second trial:

[00201] Example 20: 95%/5% blend of PP-5 with EMA-1.

[00202] Example 21: 85%/15% blend of PP-5 with EMA-1.

[00203] Spinning conditions for both trials:

[00204] Block temperature 240 °C

[00205] Melt temperature 240 °C

[00206] Spinneret 600 holes

5 [00207] Feed Roll Temperature 137 °C

[00208] Feed Roll Speed 600 rpm

[00209] Draw Roll Temperature 23 °C

[00210] Draw Roll Speed varied depending on stretching ratio (S.R.)  
desired (for example, 1800 rpm for S.R. of 3.0).

10

Table 6

Melt Blend	Comparative Example C17			Example 18		Example 19			
Maximum Stretching ratio	2.5			3.5		3.5			
Stretching ratio	1.9	2.5		1.9	2.5	3	1.9	2.5	3
Strength (g/den)	3.0	3.9		3.0	3.9	4.4	2.9	3.6	4.2
Elongation at break (%)	271	185		263	184	148	273	181	116
Linear density (den)	30	23		30	23	21	30	25	21
Hardness (g/den)	5.7	5.4		6.6	5.3	4.8	5.8	4.8	3.7

Dry Blend	Example 20				Example 21			
Maximum stretching ratio	3.5				3.5			
Stretching ratio	1.9	2.5	3	3.5	1.9	2.5	3	3.5
Strength (g/den)	2.9	3.7	4.5	3.2	2.8	3.5	4.3	5
Elongation at break (%)	246	168	135	229	254	176	136	99
Linear density (den)	29	22	22	28	30	24	20	17
Hardness (g/den)	5.2	4.8	4.8	5.4	5.2	4.7	4.5	3.8

[00211] The blends of PP and EMA-1 spun well. A significant improvement in draw ratio could be achieved with consequent tenacity increase. These trials have shown a higher stretchability (without breaks) of PP when an ethylene/alkyl acrylate copolymer is added. If for 100 % 5 PP the maximum stretching ratio was 2.5, adding 5, 10 or 15 weight % EMA-1 raised the maximum stretching ratio to 3.5. One could also see an increase in tensile strength with increased S.R. although the elongation at break is decreased.

[00212] Trial 7  
 10 [00213] The purpose of this trial was to evaluate stretchability of films prepared from unmodified and modified PP's on a tensile machine combined with an oven to be able to run the tests at elevated temperatures.

[00214] Materials used:  
 15 [00215] Comp. Example C22: 100 % PP-6 (Moplen T 30 S)  
 [00216] Comp. Example C23: 90 % PP-6 + 10 weight % Engage 8200 (a metallocene polyethylene).  
 [00217] Example 24: 95 % PP-6 + 5 weight % EMA-1  
 [00218] Example 25: 90 % PP-6 + 10 weight % EMA-1  
 20 [00219] We produced 0.5 mm thick dumbbell-shaped samples and measured their tensile properties at 100 °C. Above 100 °C, the samples did not break within the limits of the test equipment (maximum of 1500 %). The results reported in Table 7 are the average values for 20 samples per formulation, tested at 100 °C.

25

Table 7

Example	C22	C23	24	25
Tensile Strength (MPa)	26.7	19.9	21.1	22.5
Elongation at break (%)	1366	1286	1371	1440

[00220] As one can see from the above table, 10 % EMA-1 shows a 5 % increase in elongation at break vs. 100 % PP.

[00221] Biaxially oriented films

[00222] Trial 8

[00223] Samples were prepared by melt-blending polypropylene and the ethylene/alkyl acrylate copolymer and then forming a cast film by 5 extrusion. The line used to produce the biaxially oriented films had clips systems to allow for stretching in both machine and transverse directions.

[00224] Comparative Example C26: 100 % PP-7 (Basell HP 1078 polypropylene homopolymer having an MFI of 2)

[00225] Example 27: 95 weight PP-7 + 5 weight EMA-1

10 [00226] Example 28: 90 weight % PP-7 + 10 weight EMA-1

[00227] The stretching conditions were as follows:

[00228] Preheating in stretching unit: 30 or 40 seconds at 145 °C

[00229] Clip temperature: 82, 88, 100 or 105 °C

[00230] Stretching speed: 400 %/sec

15 [00231] Annealing temperature after stretching: 145 °C

Ex.	Stretch ratios	Clip Temp.	Preheat time	Tensile	Tensile	Elonga-	Elonga-
				strength MD	strength TD	tion at break MD	tion at break TD
C26	6x6	100	40	175	164	89	72
C26	6x6	100	40	169	168	91	85
27	6x6	100	40	164	178	88	99
27	6x6	100	40	176	184	89	75
28	6x6	100	40	160	152	98	106
28	6x6	100	40	161	177	88	87
28	6.5x6.5	100	40	177	176	76	83
C26	5x9	100	40	128	226	182	53
27	5x9	100	40	125	232	180	34
28	5x9	100	40	133	239	143	47
27	5x9	88	40	122	294	151	37
27	5x9	100	30	144	257	191	49
27	5x9	82	30	114	326	251	36
28	7x7	105	40	179	175	89	86
28	7x7	105	40	189	188	83	77

**[00232]** In the other trials described above, stretchability of ethylene/alkyl acrylate modified polypropylene blends using stretching rolls gave higher stretchability than 100 % PP. In this trial, stretchability

5 comparisons were difficult to make because the films were sometimes breaking at the clips, which were very small (high stress concentration) and with large spaces between them (around 10 cm). On industrial equipment, however, MD stretching is always done with rolls and not clips.

**[00233]** However, it can be observed that higher stretching of some of 10 the samples based on PP modified with EMA-1 provides better tensile strength with similar or higher elongation at break than 100 % polypropylene.

**[00234]** For the 7x7 stretched films made with 10 % EMA-1, tensile strength (average between MD and TD) was found to be 8 % higher 15 without loss in elongation at break vs. 100 % PP at 6x6 (maximum biaxial stretchability for PP).

**[00235]** For the 5x9 stretched films made with 5 % EMA-1, tensile strength (average between MD and TD) was found to be 24 % higher and elongation at break (average between MD and TD) 22 % higher vs. 100 % 20 PP at same stretching ratio of 5x9.

**[00236]** Spunbonded fabric

**[00237]** Trial 9

**[00238]** The purpose of this trial was to evaluate the effect of adding an ethylene/alkyl acrylate copolymer to polypropylene to prepare a 25 spunbonded fabric.

**[00239]** The spunbonded fabric was prepared using 2 extruders feeding a total of 68 spinning positions and laying the fibers onto a moving belt.

**[00240]** Comparative Example C29: 100 % polypropylene homopolymer PP-8 at a stretching ratio of 2.5:1.

30 **[00241]** Example 30: 95 weight % PP-8 + 5 weight % EMA-1 at a stretching ratio of 3:1.

**[00242]** Spinning conditions for the Example 30 fabric were:

**[00243]** Block temperature 240 °C

**[00244]** Melt temperature 240 °C

[00245] Feed Roll Temperature 137 °C

[00246] Feed Roll Speed 600 rpm

[00247] Draw Roll Temp 23 °C

[00248] Draw Roll Speed 1800 rpm

5 [00249] These trials have shown that Example 30 provided enhanced stretchability. The Example 30 was drawn at a 3:1 ratio, while the maximum achievable stretching ratio for Comparative Example C29 was 2.5:1. The finished fabric Example 30 has a significantly higher resistance to perforation than a standard PP nonwoven (Comparative Example C29)

10 at the same basis weight.

## CLAIMS

What is claimed is:

1. A film prepared from a composition comprising:
  - (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and
  - (b) from 1 to 30 weight % of at least one ethylene/ alkyl acrylate copolymer.
- 10 2. The film of Claim 1 wherein said alkyl acrylate is present in said ethylene/alkyl acrylate copolymer in a range from about 5 to about 30 weight %.
- 15 3. The film of Claim 1 wherein said alkyl acrylate is present in said ethylene/alkyl acrylate copolymer in a range from about 10 to about 25 weight %.
4. The film of Claim 1 wherein said alkyl acrylate is selected from the group consisting of methyl acrylate, ethyl acrylate and butyl acrylate.
- 20 5. The film of Claim 4 wherein said alkyl acrylate is methyl acrylate.
6. The film of Claim 1 wherein component (b) is present in an amount of from 2 to 30 weight %.
7. The film of Claim 6 wherein component (b) is present in an amount of from 2 to 20 weight %.
- 25 8. The film of Claim 6 wherein component (b) is present in an amount of from 2 to 10 weight %.
9. The film of Claim 1 further comprising
  - (c) from 0.01 to 40 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.
- 30 10. The film of Claim 9 wherein component (c) is present in an amount of from 0.1 to 15 weight %.
11. The film of Claim 1 that is prepared by extrusion of said composition into a cooling water bath for quenching.

12. The film of Claim 1 that is prepared by extrusion of said composition onto chilled rolls for quenching.
13. The film of Claim 1 that is prepared by extrusion of said composition through an annular die into a tubular blown film that is air-quenched.
- 5 14. The film of Claim 1 wherein said composition comprises a tubular reactor produced ethylene/alkyl acrylate copolymer.
15. A tape prepared by slitting a film of any of Claims 1 through 14.
- 10 16. A woven textile prepared from a split tape film of Claim 15.
17. An article manufactured from woven textile of Claim 16 selected from the group consisting of rope, string, cord, synthetic lawn, sacking, heavy-duty sack, primary and secondary carpet backing, geotextile, agrotextile, industrial fabric, strapping, belting, apparel, diaper, netting, container, filter, tarpaulin, awning, canopy, banner, roofing membrane, luggage liner, package, and upholstery.
- 15 18. A heavy-duty sack made of split tape film of Claim 31.
19. A geotextile made of split tape film of Claim 31.
- 20 20. An agrotextile made of split tape film of Claim 31.
21. A fiber prepared by hot-drawing and annealing a split tape of Claim 15.
- 20 22. An article manufactured from split tape of Claim 21 selected from the group consisting of rope, string, cord, synthetic lawn, sacking, heavy-duty sack, primary and secondary carpet backing, geotextile, agrotextile, industrial fabric, strapping, belting, apparel, diaper, netting, container, filter, tarpaulin, awning, canopy, banner, roofing membrane, luggage liner, package, and upholstery.
- 25 23. A process for preparing a fiber comprising
  - (1) preparing a composition comprising
  - 30 (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and

(b) from 1 to 30 weight % of at least one ethylene/alkyl acrylate copolymer;

(2) forming said composition into a film;

(3) slitting the film into tapes;

5 (4) hot-drawing and annealing a tape of step (3).

24. The process of Claim 23 wherein said alkyl acrylate is present in said ethylene/ alkyl acrylate copolymer in a range from about 5 to about 30 weight %.

25. The process of Claim 24 wherein said alkyl acrylate is 10 present in said ethylene/ alkyl acrylate copolymer in a range from about 10 to about 25 weight %.

26. The process of Claim 23 wherein said alkyl acrylate is selected from the group consisting of methyl acrylate, ethyl acrylate and butyl acrylate.

15 27. The process of Claim 26 wherein said alkyl acrylate is methyl acrylate.

28. The process of Claim 23 wherein component (b) is present in an amount from 2 to 30 weight %.

20 29. The process of Claim 28 wherein component (b) is present in an amount of from 2 to 20 weight %.

30. The process of Claim 29 wherein component (b) is present in an amount of from 2 to 10 weight %.

31. The process of Claim 23 further comprising  
25 (c) from 0.01 to 40 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.

32. The process of Claim 27 wherein component (c) is present in an amount of from 0.1 to 15 weight %.

33. The process of Claim 23 that is prepared by extrusion of said 30 composition into a cooling water bath for quenching.

34. The process of Claim 23 that is prepared by extrusion of said composition onto chilled rolls for quenching.

35. The process of Claim 23 that is prepared by extrusion of said composition through an annular die into a tubular blown film that is air-quenched.

36. The process of Claim 23 wherein said composition  
5 comprises a tubular reactor produced ethylene/alkyl acrylate copolymer.

37. A melt-spun fiber prepared from a composition comprising:  
10 (a) at least one polypropylene polymer selected from the group consisting of polypropylene homopolymers; random copolymers or block copolymers of polypropylene and ethylene; and random terpolymers or block terpolymers of polypropylene, ethylene and one other olefin; and  
(b) from 1 to 15 weight % of at least one ethylene/ alkyl acrylate copolymer.

38. The fiber of Claim 37 wherein said alkyl acrylate is present in said ethylene/ alkyl acrylate copolymer in a range from about 5 to about  
15 30 weight %.

39. The fiber of Claim 38 wherein said alkyl acrylate is present in said ethylene/ alkyl acrylate copolymer in a range from about 10 to about 25 weight %.

40. The fiber of Claim 37 wherein said alkyl acrylate is selected  
20 from the group consisting of methyl acrylate, ethyl acrylate and butyl acrylate.

41. The fiber of Claim 40 wherein said alkyl acrylate is methyl acrylate.

42. The fiber of Claim 37 wherein component (b) is present in an  
25 amount of from 2 to 10 weight %.

43. The fiber of Claim 37 further comprising  
(c) from 0.01 to 15 weight % of at least one additional component selected from the group consisting of fillers, delustrants, UV stabilizers, pigments and other additives.

30 44. The fiber of Claim 43 wherein component (c) is present in an amount of from 0.1 to 5 weight %.

45. The fiber of Claim 37 wherein said composition comprises a tubular reactor produced ethylene/alkyl acrylate copolymer.

46. A nonwoven textile prepared from a melt-spun fiber of Claim 37.
47. An article manufactured from melt-spun fiber of Claim 37 selected from the group consisting of diapers, hygiene product, medical apparel, mask, drapes, cover, blanket, packaging, durable paper, wipe, wrap, banner, carpet backing, filter, geotextile, agrotextile, industrial fabric, roofing membrane, luggage liner, upholstery, and construction wrap.

# INTERNATIONAL SEARCH REPORT

International Application No

/US2004/019658

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC 7 C08J5/18 C08L23/10 D01F6/46  
//(C08L23/10, C08L23:08)

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J C08L D01F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category <sup>o</sup>	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 614 574 A (SHETH PARESH J) 25 March 1997 (1997-03-25) claims 20,21,31,40,41 column 6, line 63 - line 66 examples	37-41,46
X	WO 95/33882 A (LYONDELL PETROCHEM) 14 December 1995 (1995-12-14) claims 1,5-8,12	37-41,46
X	WO 02/12390 A (EQUISTAR CHEM LP) 14 February 2002 (2002-02-14) claims 1-15,17-19 page 3, line 15 - line 20 page 5, line 1 - line 3 page 5, line 14 - line 19	1-9

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

<sup>o</sup> Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
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- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

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- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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## INTERNATIONAL SEARCH REPORT

ational Application No  
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 1 283 243 A (ATOFINA) 12 February 2003 (2003-02-12) claims 1,6,8 page 4, line 25 – page 7, line 6; tables -----	1
X	GB 902 809 A (SPENCER CHEMICAL COMPANY) 9 August 1962 (1962-08-09) claims 1,5-11 page 3, line 78 – line 87 -----	1-7, 37-41
X	DATABASE WPI Section Ch, Week 197803 Derwent Publications Ltd., London, GB; Class A94, AN 1978-05093A XP002295882 & JP 52 142785 A (CHUETSU POLYCOAT KK) 28 November 1977 (1977-11-28) abstract -----	1,15,21, 23,26

**INTERNATIONAL SEARCH REPORT**

International Application No

PCT/US2004/019658

Patent document cited in search report	Publication date		Patent family member(s)	Publication date
US 5614574	A 25-03-1997	US AU AU BR CA CN EP JP WO	5464687 A 695499 B2 2827295 A 9508277 A 2194238 A1 1157642 A 0770152 A1 10502687 T 9601916 A1	07-11-1995 13-08-1998 09-02-1996 28-10-1997 25-01-1996 20-08-1997 02-05-1997 10-03-1998 25-01-1996
WO 9533882	A 14-12-1995	US US AU AU BR CA CN EP JP WO	5550192 A 5576366 A 690997 B2 2603395 A 9507945 A 2190376 A1 1157015 A 0770156 A1 10501309 T 9533882 A1	27-08-1996 19-11-1996 07-05-1998 04-01-1996 02-09-1997 14-12-1995 13-08-1997 02-05-1997 03-02-1998 14-12-1995
WO 0212390	A 14-02-2002	US AU WO	6433088 B1 8075101 A 0212390 A2	13-08-2002 18-02-2002 14-02-2002
EP 1283243	A 12-02-2003	FR CA CN EP JP US	2828493 A1 2395685 A1 1401689 A 1283243 A1 2003105145 A 2003130429 A1	14-02-2003 07-02-2003 12-03-2003 12-02-2003 09-04-2003 10-07-2003
GB 902809	A 09-08-1962	BE DE FR NL NL	606041 A 1197226 B 1294699 A 126298 C 266984 A	22-07-1965 26-05-1962
JP 52142785	A 28-11-1977	JP JP	1100562 C 55025056 B	18-06-1982 03-07-1980