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54 **Biconstituent polypropylene/polyethylene fibers.**

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## Description

Blends consisting of polypropylene and polyethylene are spun into fibers having improved properties.

Polypropylene (PP) fibers and filaments are items of commerce and have been used in making products such as ropes, non-woven fabrics, and woven fabrics.

US-A-4,578,414 discloses additives for making olefin polymer fibers water-wettable, including blends of polyethylene (PE) and polypropylene (PP).

US-A-4,518,744 discloses melt-spinning of certain polymers and blends of polymers, including polypropylene (PP). JP-A-56-159339 and JP-A-56-159340 disclose fibers of mixtures of polyester with minor amounts of polypropylene.

Convenient references relating to fibers and filaments, including those of man-made thermoplastics, and incorporated herein by reference, are, for example:

(a) Encyclopedia of Polymer Science and Technology, Interscience, New York, Vol. 6 (1967), pp. 505-555 and Vol. 9 (1968), pp. 403-440;

(b) Man-Made Fiber and Textile Dictionary, published by Celanese Corporation;

(c) Fundamentals of Fibre Formation--The Science of Fibre Spinning and Drawing, by Andrzej Ziabicki published by John Wiley & Sons, London/New York, 1976;

(d) Man-Made Fibres, by R. W. Moncrieff, published by John Wiley & Sons, London/New York, 1975;

(e) Kirk-Othmer Encyclopedia of Chemical Technology, Vol. 16 for "Olefin Fibers", published by John Wiley & Sons, New York, 1981, 3rd Edition.

In conformity with commonly accepted vernacular or jargon of the fiber and filament industry, the following definitions apply to the terms used in this disclosure:

A "monofilament" (also known as (a.k.a.) monofil) refers to an individual strand of denier greater than 15 (1.7 tex), usually greater than 30 (3.3 tex);

A "fine denier fiber or filament" refers to a strand of denier less than 15 (1.7 tex);

A "multi-filament" (a.k.a. multifil) refers to simultaneously formed fine denier filaments spun as a bundle of fibers, generally containing at least 3, preferably at least 15-100 fibers and can be several hundred or several thousand;

"Staple fibers" refer to fine denier strands which have been formed at, or cut to, staple lengths of generally 1 to 8 inches (2.5 to 20 cm);

An "extruded strand" refers to an extrudate formed by passing polymer through a forming-orifice, such as a die.

A "fibril" refers to a superfine discrete filament embedded in a more or less continuous matrix.

Whereas it is known that virtually any thermoplastic polymer can be extruded as a coarse strand or monofilament, many of these, such as polyethylene and some ethylene copolymers, have not generally been found to be suitable for the making of fine denier fibers or multi-filaments. Practitioners are aware that it is easier to make a coarse monofilament yarn of 15 denier (1.7 tex) than to make a multi-filament yarn of 15 denier (1.7 tex). It is also recognized that the mechanical and thermal conditions experienced by a bundle of filaments, whether in spinning staple fibers or in multi-filaments yarns, are very different to those in spinning monofilaments. The fact that a given man-made polymer can be extruded as a monofilament, does not necessarily herald its use in fine denier (tex) or multi-filament spinning. Whereas an extruded monofilament which has been cooled can usually be cold-drawn (stretched) to a finer denier size, even if it does not have sufficient melt-strength to be melt-drawn without breaking, it is apparent that a polymer needs to have an appreciable melt-strength to be hot-drawn to fine denier sizes.

Low density polyethylene (LDPE) is prepared by polymerizing ethylene using a free-radical initiator, e.g. peroxide, at elevated pressures and temperatures, having densities in the range, generally, of 0.910-0.935 g/cm<sup>3</sup>. The LDPE, sometimes called "I.C.I.-type" polyethylene is a branched (i.e. non-linear) polymer, due to the presence of short-chains of polymerized ethylene units pendent from the main polymer backbone. Some of the older art refers to these as high pressure polyethylene (HPPE).

High density polyethylene (HDPE) is prepared using a coordination catalyst, such as a "Ziegler-type" or "Natta-type" or a "Phillips-type" chromium oxide compound. These have densities generally in the range of 0.94 to 0.98 g/cm<sup>3</sup> and are called "linear" polymers due to the substantial absence of short polymer chains pendent from the main polymer backbone.

Linear low density polyethylene (LLDPE) is prepared by copolymerizing ethylene with at least one  $\alpha$ -olefin alkylene of C<sub>3</sub>-C<sub>12</sub>, especially at least one of C<sub>4</sub>-C<sub>8</sub>, using a coordination catalyst such as is used in making HDPE. These LLDPE are "linear", but with alkyl groups of the  $\alpha$ -olefin pendent from the polymer chain. These pendent alkyl groups cause the density to be in about the same density range (0.88-0.94 g/cm<sup>3</sup>) as the LDPE; thus the name "linear low density polyethylene" or LLDPE is used in the industry in

referring to these linear low density copolymers of ethylene.

Polypropylene (PP) is known to exist as atactic (largely amorphous), syndiotactic (largely crystalline), and isotactic (also largely crystalline), some of which can be processed into fine denier fibers. It is preferable, in the present invention, to use the largely crystalline types of PP suitable for spinning fine denier fibers, sometimes referred to as "CR", or constant rheology, grades.

US-A-4,181,762, US-A-4,258,097, and US-A-4,356,220 contain information about olefin polymer fibers, some of which are monofilaments.

US-A-4,076,698 discloses methods of producing LLDPE and discloses extrusion of a monofilament.

US-A-4,584,347 discloses in general terms the manufacture of xerogel fibers from dilute solutions of ultra high molecular weight polyethylene or polypropylene containing a polymeric additive which can be LDPE, LLDPE or HDPE but there is no exemplification of a PE/PP mix.

US-A-4,563,504 discloses the manufacture of mono-oriented yarns from a mixture of 10-40 weight percent polypropylene and 60 to 90 weight percent ethylene  $\alpha$ -olefin copolymer. The components can be mixed in the solid or molten state.

US-A-4,632,861 discloses that the melt spinning of LDPE is improved by blending LDPE with polypropylene in the amounts of 65-95 weight percent LDPE and 5 to 35 weight percent PP. The resultant fibers have PP dispersed in a PE continuous phase. Comparative examples having 40:60 and 20:80 LDPE:PP are given but the patent teaches that PP content below 35 weight percent is required for satisfactory spinning.

CA-A-1199746 discloses mixtures of 40 to 90 weight percent LLDPE to improve the flexibility, capacity for hot and cold drawing, and strength of PP for blow-molding, extrusion drawing or thermoforming. No reference is made to spinning fibers.

Skoroszewski ("Parameters Affecting Processing of Polymers and Polymer Blends", *Plastics & Polymers*, Volume 40, No. 147, pages 142-152, July 1972) discloses stretched polypropylene film fibers containing LDPE and teaches that LDPE contents above 20 weight percent drastically affect the tenacity of the products.

JP-A-52072744, JP-A-58011536, JP-A-58206647 and JP-A-5904132 disclose moulding compositions comprising polypropylene and polyethylene but make no reference to fiber manufacture. JP-A-52072744 discloses compositions containing 70 to 98 weight percent polypropylene, 1 to 14 weight percent ethylene/but-1-ene random copolymers and 1 to 15 weight percent LDPE. JP-A-58011536 discloses compositions comprising an ethylene propylene random copolymer, containing 3 to 9 weight percent ethylene, and a LLDPE. JP-A-58206647 discloses a composition comprising polypropylene, LDPE and LLDPE. JP-A-59041342 discloses compositions containing 60 to 95 weight percent LLDPE and 5 to 50 weight percent polypropylene.

DE-A-3544523 discloses the production of biconstituent fibers by a process in which a finely dispersed mixture of polypropylene and 5 to 20 wt. %, preferably 10 to 20 wt. %, polyethylene is melt spun. The biconstituent fibers have higher polyethylene density near the fiber surface, where the polyethylene fibrils are axially oriented. The polyethylene can be LLDPE.

It has now been found, unexpectedly, that improvements are made in polypropylene fibers if the polypropylene is first blended with 20 percent to 45 percent by weight of a linear low density ethylene copolymer and the molten mixture intimately mixed immediately prior to melt spinning.

According to a first aspect, the invention provides a biconstituent fiber consisting essentially of polypropylene as a continuous phase, having distributed therein 20 to 45 percent by weight of linear low density polyethylene (LLDPE) fibrils as a dispersed phase arrayed in a substantially omni-directionally splayed manner, said LLDPE having a melt flow rate (as measured in accordance with ASTM D-1238 (E)) in the range of 12 to 120 g/10 min. The tenacity and softness of the fibers is improved over that of the polypropylene or the polyethylene alone.

According to a second aspect of the invention, there is provided a process of preparing a biconstituent fibers which comprises intimately mixing molten polypropylene (PP) and molten linear low density polyethylene (LLDPE), having a melt flow rate (as measured in accordance with ASTM D-1238 (E)) in the range 12 to 120 g/10 min in the PP:LLDPE weight ratio 80:20 to 55:45 to disperse the LLDPE in the PP and maintaining the dispersion until the mixture, as an extrudate, is expelled from a spinning die to form a fiber in which LLDPE fibrils, as a dispersed phase, are arrayed in a substantially omni-directional splayed manner.

## IN THE DRAWINGS

Figs. 1-4 are provided herewith as visual aids for relating certain properties of blends described in this

disclosure.

The polyethylene for use in this invention is LLDPE with a molecular weight of the polyethylene in the moderately high range, as indicated by a melt index, M.I., (a.k.a. melt flow rate, M.F.R.) value in the range of from 12 to 120, preferably 20 to 100 g/10 min, especially  $50 \pm 20$  g/10 mins, as measured by ASTM D-1238(E) (190° C/2.16 Kg).

It is preferred that the comonomer  $\alpha$ -olefin alkylenes in the LLDPE are, in the C<sub>3</sub>-C<sub>12</sub> range, especially C<sub>4</sub> to C<sub>8</sub> and particularly 1-octene. Butene (C<sub>4</sub>) can be used, but 1-octene is preferred. Mixtures of the alkylene comonomers may be used, such as butene/octene or hexene/octene in preparing the ethylene/alkylene copolymers. The density of the LLDPE is dependent on the amount of, and the molecular size (i.e. the number of carbons in the alkylene molecule) of, the alkylene incorporated into the copolymer. The more alkylene comonomer used, the lower the density; also, the larger the alkylene comonomer, the lower the density. Preferably an amount of alkylene comonomer is used which results in a density in the range of 0.88 to 0.94, most preferably 0.92 to 0.94, especially 0.92 to 0.93 g/cm<sup>3</sup>. An ethylene/octene copolymer having a density of around 0.925 g/cm<sup>3</sup>, an octene content in the range of from 5 to 10 percent and a M.F.R. of  $50 \pm 20$  g/10 min. is very effective for the purposes of this invention.

In the blend, the weight ratio of PP/PE can range from 80/20 to 55/45, but is preferably in the range of 78/22 to 60/40, most preferably in the range of 75/25 to 65/35. An especially preferred range is 72/28 to 68/32.

The method of melt-mixing is important due to generally acknowledged immiscibility of the PP and PE. An intensive mixer-extruder is required which causes, in the blender, on the one hand, molten PE to be dispersed in the molten PP and the dispersion maintained until the mixture, as an extrudate, is expelled from the spinning die.

The following chart is provided as a means for describing the results believed to be obtained for the various ratio ranges of PP/PE, when using PE having an M.F.R. in the range of 12 to 120 g/10 min., and a crystalline PP, where the melt viscosity and melt strength are such that reasonably good melt-compatibility and miscibility are achieved by use of the high-intensity mixer-extruder:

Approx. Range of Ratio of PE/PP	General results one may obtain*
20/80 - 45/55	PE fibrils dispersed in PP continuous matrix
45/55 - 55/45	co-continuous zones; lamellar structure
55/45 - 90/10	PP fibrils dispersed in PE continuous matrix

\*Obviously the results in or around the ratios which are overlapping at the ends of the middle range are ambiguous in that some of results obtained from both sides of the overlap.

Polymer blends of PP and PE prepared in such a mixer are found to be useful, strong, and can be extruded into products where the immiscibility is not a problem. As the so-formed extrudate of a mixture which contains more PP than PE is spun and drawn into fibers, the molten PE globules become extruded into fibrils within the polypropylene matrix. An important, novel feature of the fibers is that the fibrils of PE are diverse in their orientation in the PP fiber. A larger fraction of PE particles is found close to the periphery of the cross-section of the PP fibers, and the remaining PE particles are spread in the inner portions of the PP fiber. The size of the PE particles is smallest at the periphery of the fiber's cross-section and a gradual increase in size is evidenced toward the center of the fiber. The frequency of small particles at the periphery is highest, and it decreases toward the center where the PE particles are largest, but spread apart more. The PE fibrils near the periphery of the PP fiber's cross-section are diverse in the direction in which they are oriented or splayed, whereas close to the center of the PP fiber the orientation is mostly coaxial with the fiber. For the purpose of being concise, these fibers will be referred to herein as blends consisting of PP as a continuous phase, and containing omni-directionally splayed PE fibrils as a dispersed phase.

Microscopic examination reveals that the PE fibrils, when viewed in a cross-section of the biconstituent PP fiber, are more heavily populated near the outer surface than in the middle. The shape of each PE fibril in the cross-section is dependent on whether one is viewing a PE fibril sliced at right angles to the axis of the PE fibril at that point or at a slant to the axis of the PE fibril at that point. An oval or elongate shaped section indicates a PE fibril cut at an angle. An elongate shaped section indicates a PE fibril which has skewed from axial alignment to a transverse position.

The mixer for preparing the molten blend of PP/PE preferably is a dynamic high intensity mixer, especially one which provides 3-dimensional mixing. Insufficient mixing will cause non-homogeneous dispersion of PE in PP resulting in fibers of inconsistent properties, and tenacities lower than that of the corresponding PP fibers alone. A 3-dimensional mixer suitable for use in the present invention is disclosed in a publication titled "Polypropylene--Fibers and Filament Yarn With Higher Tenacity", presented at International Man-Made Fibres Congress, September 25-27, 1985, Dornbirn/Austria, by Dr. Ing. Klaus Schafer of Barmag, Barmer Maschinen-Fabrik, West Germany.

The distribution of PE fibrils in a PP matrix are studied by using the following method: The fibers are prepared for transverse sectioning by being attached to strips of adhesive tape and embedded in epoxy resin. The epoxy blocks are trimmed and faced with a glass knife on a Sorvall MT-6000 microtome. The blocks are soaked in a mixture of 0.2 gm ruthenium chloride dissolved in 10 ml of 5.25 percent by weight aqueous sodium hypochlorite for 3 hours. This stains the ends of the fibers with ruthenium to a depth of about 30 micrometers. The blocks are rinsed well and remounted on the microtome. Transverse sections of fibers in epoxy are microtomed using a diamond knife, floated onto a water trough, and collected onto copper TEM grids. The grids are examined at 100 KV accelerating voltage on a JEOL 100C transmission electron microscope (TEM). Sections taken from the first few micrometers, as well as approximately 20 micrometers from the end are examined in the TEM at magnifications of 250X to 66,000X. The polyethylene component in the samples are preferentially stained by the ruthenium. Fiber sections microtomed near the end of the epoxy block may be overstained, whereas sections taken about 20 micrometers away from the end of the fibers are more likely to be properly stained. Scratches made by the microtome knife across the face of the section may also contain artifacts of the stain, but a skilled operator can distinguish the artifacts from the stained PE. The diameter of PE fibrils near the center of the PP fiber have been found to be, typically, on the order of about 350 to 500 angstrom (35 to 50 nm), whereas the diameter of the more populace fibrils near the periphery edge of the PP fiber have been found to be, typically, on the order of about 100 to 200 angstrom (10 to 20 nm). This is in reference to those which appear under high magnification to be of circular cross-section rather than oval or elongate.

At less than 20 percent polyethylene in the polypropylene one obtains better "hand" than with polypropylene alone, but without obtaining a significant increase in tenacity and without obtaining a dimensionally stable fiber. By the term "dimensionally stable" it is meant that upon storing a measured fiber for several months and then remeasuring the tenacity, one does not encounter a significant change in the tenacity. A change in tenacity indicates that stress relaxation has occurred and that fiber shrinkage has taken place. In many applications, such as in non-woven fabrics, such shrinkage is considered undesirable.

By using 20 percent to 45 percent polyethylene in the polypropylene one obtains increased tenacity as well as obtaining better "hand" than with polypropylene alone. By using between 25 percent to 35 percent, especially 28 percent to 32 percent, of polyethylene in the polypropylene one also obtains a substantially dimensionally stable fiber. A substantially dimensionally stable fiber is one which undergoes very little, if any, change in tenacity during storage. A ratio of polypropylene/polyethylene of 70/30 is especially beneficial in obtaining a dimensionally stable fiber. By using 50 percent to 90 percent polyethylene in the blend, a reduction in tenacity may be observed, but the "hand" is noticeably softer than polypropylene alone.

A greater draw ratio gives a higher tenacity than a lower draw ratio. Thus, for a given PP/PE ratio, a draw ratio of, say 3.0 may yield a tenacity greater than PP alone, but a draw ratio of, say 2.0 may not give a greater tenacity than PP alone.

In order to establish a nominal base point for making comparisons, several commercially available PP's are spun into fine denier fibers and the results are averaged. The average denier size is found to be 2.1 (0.23 tex), the average elongation is found to be 208 percent and the average tenacity at the break point is 2.26 g/denier (0.199 N/tex).

Similarly, to establish a nominal base point, several LLDPE samples are spun into fine denier fibers and the results are averaged. The average denier size is found to be 2.84, the average elongation is found to be 141 percent, and the average tenacity at the break point is 2.23 g/denier (0.197 N/tex).

The following examples illustrate particular embodiments, but the invention is not limited to these particular embodiments.

EXAMPLE 1

A blend of 80 percent by weight of PP granules (M.I., 230° C/2.16 kg, about 25 g/10 min. and density of 0.910 g/cc) with 20 percent by weight of LLDPE (1-octene; M.I. of 50 g/10 min.; density of 0.926 g/cm<sup>3</sup>) is mechanically mixed and fed into an extruder maintained at about 245 to 250° C where the polymers are melted. The molten polymers are passed through a 3-dimensional dynamic mixer mounted at the outlet of the extruder. The dynamic mixer is designed, through a combination of shearing and mixing, to simultaneously divide the melt stream into superfine layers, and rearrange the layers tangentially, radially, and axially, thereby effecting good mixing of the immiscible PP and LLDPE.

The so-mixed melt is transported from the dynamic mixer, by a gear pump, through a spinneret having 20,500 openings. The formed filaments are cooled by a side-stream of air, wound on a take-up roller, stretched over a preheated heptet of Godet rollers (90° to 140° C), run through an air-heated annealing oven (150° to 170° C), followed by another heptet of Godet rollers (100° to 140° C), before crimping and cutting of the continuous fibers into 38 mm staple fibers. Appropriate spin-finishes are applied to aid the operation. The stretch ratio is 3.1X.

The resulting fibers have about 20 cpi (crimps per inch) (8 crimps per cm) and the titre is in the range of 2.0-2.5 dpf (denier per filament) (0.22 - 0.28 tex per filament). The mechanical properties of the fibers, measured 3 weeks after production, are as follows (average of 15 randomly sampled fibers): Titre of 2.14 dpf (0.238 tex per filament); tenacity (tensile at break) of 4.73 g/denier (0.417 N/tex); elongation (at break) of 52 percent. The "hand" (softness) was judged better than that of similar PP fibers alone.

EXAMPLE 2

This example is like Example 1 above except that 30 weight percent of the LLDPE and 70 weight percent of the PP is used.

Results: Titre of 2.66 dpf (0.296 tpf (tex per filament)); tenacity of 3.23 g/denier (0.285 N/tex); elongation of 61 percent. The hand was clearly better than PP alone.

EXAMPLE 3

This example is like Example 1 above except that the LLDPE contains 1-butene instead of 1-octene. It also has M.I. of 50 g/10 min., a density of 0.926 g/cm<sup>3</sup>, and comprises 20 percent by weight of the blend.

Results: Titre of 2.24 dpf (0.249 tpf); tenacity of 3.93 g/denier (0.347 N/tex); elongation of 48 percent. The hand was judged better than PP alone.

Table IA below illustrates the change in properties when measured about 120 days following the initial measurements shown in Examples 1-3 above.

TABLE IA

Example	Ratio PP/PE	DENIER		TENACITY (g/denier) (N/tex)		ELONGATION (%)	
		First Measure	Second Measure	First Measure	Second Measure	First Measure	Second Measure
1	80/20	2.14 (0.238)	2.81 (0.312)	4.73 (0.417)	3.41 (0.301)	52	70
2	70/30	2.66 (0.296)	2.69 (0.299)	3.23 (0.285)	3.37 (0.297)	61	72
3	80/20	2.24 (0.249)	3.00 (0.333)	3.93 (0.347)	2.99 (0.264)	48	63

The fibers of Examples 1 and 2 were evaluated again after the exposure to 60° C for 42 days plus balance to 14 months at room temp (25° C), and the results were obtained as shown in Table IB.

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TABLE IB

EXAMPLE	DENIER (tex)	TENACITY (g/denier) ((N/tex))	ELONGATION (%)
1	3.07 (0.341)	3.42 (0.302)	41.0
2	3.11 (0.346)	3.20 (0.282)	41.6

The 70/30 blend (Example 2) in the tables above exhibited very little change in tenacity; this is an indication that these particular biconstituent fibers show unusual permanency of strength, affected very little by stress relaxation during storage. The 70/30 blend is found to form a high strength non-woven structure (about 2650 gm. force (26.0 N) to break a 1 inch (2.5 cm) wide strip) when thermally bonded at about 148° C under 700 psi (4.8 MPa) pressure to form a 1 oz./yd<sup>2</sup> (34 g/m<sup>2</sup>) fabric.

EXAMPLE 4

In this set of data, the following described blends are used, wherein the PP used in each is a highly crystalline PP having a M.F.R. of 25 g/10 minutes as measured by ASTM D-1238 (230° C, 2.16 Kg) and the M.F.R. of the PE's are measured by ASTM D-1238 (190° C, 2.16 Kg). All of the PE's are LLDPE's identified as:

- PE-A - LLDPE (1-octene comonomer), 50 M.F.R., 0.926 density
- PE-B - LLDPE (1-octene comonomer), 105 M.F.R., 0.930 density
- PE-C - LLDPE (1-octene comonomer), 26 M.F.R., 0.940 density
- PE-D - LLDPE (1-butene comonomer), 50 M.F.R., 0.926 density

Blends made of the above described polymers are made into fibers in the manner described hereinbefore, the results of which are shown below in Table II.

TABLE II

Run No.	PE Used	Wt. Ratio PE/PP	Stretch Ratio	Titer (denier) (tex)	Tenacity g/denier (N/tex)	% Elong.	
5	1	A	25/75	2.0	4.15 (0.461)	1.87 (0.165)	191
	2	A	25/75	2.7	2.88 (0.320)	2.61 (0.230)	99
	3	A	45/55	2.0	4.15 (0.461)	1.67 (0.147)	217
	4	A	45/55	2.85	3.27 (0.363)	2.17 (0.192)	140
10	5	A	65/35	2.0	4.79 (0.532)	1.13 (0.100)	298
	6	A	65/35	2.7	3.53 (0.392)	1.56 (0.138)	208
	7	A	85/15	2.0	4.27 (0.474)	1.00 (0.088)	307
	8	A	85/15	2.7	3.52 (0.391)	1.21 (0.107)	216
	9	A	85/15	3.0	3.06 (0.340)	1.63 (0.144)	150
15	10	B	25/75	2.0	4.48 (0.498)	1.88 (0.166)	243
	11	B	25/75	3.1	2.88 (0.320)	2.85 (0.252)	76
	12	B	45/55	2.0	4.23 (0.470)	1.47 (0.130)	225
	13	B	45/55	3.1	2.85 (0.317)	2.18 (0.192)	100
	14	B	65/35	2.0	4.17 (0.463)	1.07 (0.094)	261
20	15	B	65/35	3.1	2.65 (0.294)	1.74 (0.154)	113
	16	D	25/75	2.0	3.87 (0.430)	1.96 (0.173)	199
	17	D	25/75	2.7	2.91 (0.323)	2.87 (0.253)	84
	18	D	25/75	3.1	2.51 (0.279)	3.61 (0.319)	41
	19	D	45/55	2.0	4.15 (0.461)	1.62 (0.143)	241
25	20	D	45/55	2.7	3.07 (0.341)	2.06 (0.182)	126
	21	D	65/35	2.0	4.39 (0.488)	1.01 (0.089)	291
	22	D	65/35	2.7	3.08 (0.342)	1.50 (0.132)	145
	23	C	25/75	2.0	3.95 (0.439)	2.11 (0.186)	219
	24	C	25/75	3.1	2.66 (0.296)	3.17 (0.280)	80
30	25	C	25/75	3.5	2.36 (0.262)	3.06 (0.270)	91
	26	C	25/75	2.3	2.64 (0.293)	2.73 (0.241)	81
	27	C	25/75	2.3	2.11 (0.234)	2.46 (0.217)	144
	28	C	45/55	2.0	4.01 (0.446)	1.90 (0.168)	266
	29	C	45/55	3.1	2.72 (0.302)	3.43 (0.303)	76
35	30	C	45/55	3.5	2.05 (0.228)	3.64 (0.321)	50
	31	C	45/55	2.7	2.88 (0.320)	3.08 (0.272)	80
	32	C	65/35	2.0	4.12 (0.458)	1.54 (0.136)	321
	33	C	65/35	2.7	3.05 (0.339)	2.19 (0.193)	169
	34	C	85/15	2.0	3.94 (0.438)	1.28 (0.113)	351
40	35	C	85/15	2.7	2.84 (0.316)	1.83 (0.162)	194
	36	C	85/15	3.1	2.79 (0.310)	2.01 (0.177)	187

Fig. 1 illustrates some of the data for PE-A.

Fig. 2 illustrates some of the data for PE-B.

Fig. 3 illustrates some of the data for PE-C.

Fig. 4 illustrates some of the data for PE-D.

Thermal bondability of biconstituent fibers are demonstrated using a PE/PP blend of 30/70 wherein PE-A is employed. After being stored for 150 days after spinning, thermal bonding is tested by preparing 10 samples of 1 inch (2.5 cm) wide slivers using a rotaring device, such as is commonly used in the industry, aiming at 1 oz. per yd<sup>2</sup> (34 g/m<sup>2</sup>) web weight. Results of the 10 measurements are normalized to 1 oz. per yd<sup>2</sup> (34 g/m<sup>2</sup>). The pressure between the calanders during the thermal bonding is maintained constant at 700 psig (4.8 MPa) in preparing fabrics. Listed below are the bonding temperature and corresponding tensile force, in grams, required to break the fabric.

	<u>Bonding Temp. °C</u>	<u>Force to Break, Grams (N)</u>
5	141	1260 (12.4)
	144	1250 (12.3)
	147	2600 (25.5)
10	149	2750 (27.0)

For comparison with the above, the typical break force usually obtained for PP base fabrics is 2500±150 grams and the typical range usually obtained for LLDPE is 1300-1500 grams.

15 It is noticed that the "drape" and softness of fabrics made using the PE/PP biconstituent fibers in spun-bonding is superior to that of PP fibers alone.

In similar manner, fibers are prepared using a melt temperature in the range of 180° to 260° C, preferably 200° to 250° C. Spinning rates of 20 to 150 m/min. are preferred. Stretch ratios in the range of 1.5-5X, preferably 2.0-3.0X are preferred. At excessive Godet rolls temperatures, sticking of the fibers to the  
20 rolls may take place. A proper choice of a spinline would tend to alleviate or minimize this, within a reasonable temperature range.

Practitioners of the art routinely measure the "hand" (softness) by merely feeling and squeezing a wad or mat of the fibers being compared.

25 The diameter of the PE fibrils which are contained in the fibers are all of sub-micrometer size and most of them have a diameter of less than about 0.05 micrometers.

Whereas the fibers may be of any denier size, the preferred denier size is less than 30 (3.3 tex) and the most preferred denier size is in the fine denier range of 0.5 to 15 (0.05 to 1.7 tex), especially in the range of 1 to 5 (0.1 to 0.6 tex).

30 The fibers of this invention are useful in a variety of applications, such as non-wovens, wovens, yarns, ropes, continuous fibers, and fabrics such as carpets, upholstery, wearing apparel, tents, and industrial applications such as filters and membranes.

The blends over the range of PP/PE ratios of 20/80 to 90/10 exhibit surprisingly good strength during extrusion and are not subject to the breaking one normally obtains from blends of incompatible polymers.

### 35 Claims

1. A biconstituent fiber consisting essentially of polypropylene as a continuous phase, having distributed therein linear low density polyethylene (LLDPE) fibrils as a dispersed phase characterized in that said LLDPE has a melt flow rate (as measured in accordance with ASTM D-1238 (E)) in the range of 12 to  
40 120 g/10 min and constitutes 20 to 45 percent by weight of the fiber and said fibrils thereof are arrayed in a substantially omni-directionally splayed manner.
2. A fiber as claimed in Claim 1, wherein said LLDPE has a density in the range of 0.92 to 0.94 g/cm<sup>3</sup>.
- 45 3. A fiber as claimed in Claim 1 or 2, wherein said LLDPE has an alkylene comonomer content in the range of 3 to 20 percent by weight of the LLDPE.
4. A fiber as claimed in any one of the preceding claims, wherein the fiber has a size of less than 3.3 tex (30 denier).
- 50 5. A fiber as claimed in Claim 4, wherein the fiber has a size of 0.05 to 1.7 tex (0.5-15 denier).
6. A fiber as claimed in Claim 5, wherein the fiber has a size in the range of 0.1 to 0.6 tex (1 to 5 denier).
- 55 7. A fiber as claimed in any one of the preceding claims, wherein the LLDPE has a melt flow rate of 20 to 100 g/10 min.
8. A fiber as claimed in Claim 7, wherein the melt flow rate is 50 ± 20 g/10 min.

9. A fiber as claimed in any one of the preceding claims, wherein the LLDPE has a density of 0.92 to 0.93 g/cm<sup>3</sup>.
10. A fiber as claimed in any one of the preceding claims, wherein the alkylene comonomer contains 4 to 8 carbon atoms.
11. A fiber as claimed in any one of the preceding claims, wherein the polyethylene comprises 25 to 35 weight percent of the total.
12. A fiber as claimed in Claim 11, wherein the polyethylene comprises 28 to 32 weight percent of the total.
13. A fiber as claimed in any one of the preceding claims, wherein the polyethylene fibrils are of sub-micrometer size and predominantly of a diameter of less than 0.05 micrometers.
14. A fiber as claimed in any one of the preceding claims, wherein the said alkylene comonomer is 1-octene.
15. A fiber as claimed in Claim 14, wherein the LLDPE has a density of about 0.925 g/cm<sup>3</sup>, an octene content of 5 to 10 percent, and a melt flow index of 50 ± 20 g/10 min.
16. A process of preparing biconstituent fibers which comprises expelling from a spinning die an intimate mixture of molten polypropylene (PP) and molten linear low density polyethylene (LLDPE), characterized in that said LLDPE has a melt flow rate (as measured in accordance with ASTM D-1238 (E)) in the range of 12 to 120 g/10 min and the PP:LLDPE weight ratio is 80:20 to 55:45 whereby the fibers have LLDPE fibrils, as a dispersed phase, arrayed in a substantially omni-directionally splayed manner.
17. A process as claimed in Claim 16, wherein the mixing is conducted using a 3-dimensional dynamic mixer.
18. A process as claimed in Claim 16 or Claim 17, wherein the components of the mixture are as specified in any one of Claims 2, 3, 7 to 12, 14 and 15 and/or the mixture is spun to provide fibers as claimed in any one of Claims 4 to 6 and 13.

#### Patentansprüche

1. Zweikomponentenfaser, bestehend im wesentlichen aus Polypropylen als kontinuierlicher Phase, in welcher Fibrillen aus linearem Polyethylen niedriger Dichte (LLDPE) als eine dispergierte Phase verteilt sind, dadurch gekennzeichnet, daß dieses LLDPE einen Schmelzindex (gemessen nach der Norm ASTM D-1238 (E)) im Bereich von 12 bis 120 g/10 min besitzt und 20 bis 45 Gew.-% der Faser ausmacht, und daß diese Fibrillen hieraus in einer im wesentlichen ungerichtet ausgebreiteten Weise angeordnet sind.
2. Faser nach Anspruch 1, worin dieses LLDPE eine Dichte im Bereich von 0,92 bis 0,94 g/cm<sup>3</sup> besitzt.
3. Faser nach Anspruch 1 oder 2, worin dieses LLDPE einen Alkylencomonomergehalt im Bereich von 3 bis 20 Gew.-% des LLDPE besitzt.
4. Faser nach einem der vorhergehenden Ansprüche, worin die Faser eine Größe von weniger als 3,3 tex (30 Denier) besitzt.
5. Faser nach Anspruch 4, worin die Faser eine Größe im Bereich von 0,05 bis 1,7 tex (0,5 - 15 Denier) besitzt.
6. Faser nach Anspruch 5, worin die Faser eine Größe im Bereich von 0,1 bis 0,6 tex (1 bis 5 Denier) besitzt.
7. Faser nach einem der vorhergehenden Ansprüche, worin das LLDPE eine Schmelzindex von 20 bis 100

g/10 min besitzt.

8. Faser nach Anspruch 7, worin der Schmelzindex  $50 \pm 20$  g/10 min beträgt.
- 5 9. Faser nach einem der vorhergehenden Ansprüche, worin das LLDPE eine Dichte von 0,92 bis 0,93 g/cm<sup>3</sup> besitzt.
10. Faser nach einem der vorhergehenden Ansprüche, worin das Alkylencomonmere 4 bis 8 Kohlenstoffatome enthält.
- 10 11. Faser nach einem der vorhergehenden Ansprüche, worin das Polyethylen 25 bis 35 Gew.-% der Gesamtmenge ausmacht.
12. Faser nach Anspruch 11, worin das Polyethylen 28 bis 32 Gew.-% der Gesamtmenge ausmacht.
- 15 13. Faser nach einem der vorhergehenden Ansprüche, worin die Polyethylenfibrillen von Submikrometer-Abmessung sind und überwiegend von einem Durchmesser von weniger als 0,05  $\mu\text{m}$ .
14. Faser nach einem der vorhergehenden Ansprüche, worin dieses Alkylencomonmere 1-Okten ist.
- 20 15. Faser nach Anspruch 14, worin das LLDPE eine Dichte von etwa 0,925 g/cm<sup>3</sup>, einen Oktengehalt von 5 bis 10 % und einen Schmelzindex von  $50 \pm 20$  g/10 min besitzt.
- 25 16. Verfahren zur Herstellung von Zweikomponentenfasern, welches das Auspressen einer innigen Mischung aus geschmolzenem Polypropylen (PP) und linearem Polyethylen niedriger Dichte (LLDPE) aus einem Spinnwerkzeug umfaßt, dadurch gekennzeichnet, daß dieses LLDPE einen Schmelzindex (gemessen nach der Norm ASTM D-1238(E)) im Bereich von 12 bis 120 g/10 min besitzt, und daß das Gewichtsverhältnis von PP:LLDPE 80:20 bis 55:45 beträgt, wodurch die Fasern LLDPE-Fibrillen als dispergierte Phase, in im wesentlichen ungerichtet ausgebreiteter Weise angeordnet, aufweisen.
- 30 17. Verfahren nach Anspruch 16, worin das Mischen unter Verwendung eines dreidimensionalen, dynamischen Mixers durchgeführt wird.
- 35 18. Verfahren nach Anspruch 16 oder Anspruch 17, worin die Bestandteile der Mischung, wie in einem der Ansprüche 2, 3, 7 bis 12, 14 und 15 angegeben, sind, und/oder die Mischung zur Lieferung von Fasern, wie in einem der Ansprüche 4 bis 6 und 13 angegeben, versponnen wird.

### Revendications

- 40 1. Fibre à deux composants essentiellement constituée de polypropylène en tant que phase continue, comportant, réparties dans celle-ci, des fibrilles de polyéthylène basse densité linéaire (PEbdl) en tant que phase dispersée, caractérisée en ce que ledit PEbdl a un indice de fluidité à chaud [mesuré selon ASTM D-1238) (E)] dans la plage allant de 12 à 120 g/10 min et constitue de 20 à 45 % en poids de la fibre, et lesdites fibrilles de celle-ci sont disposées de façon à s'étendre pratiquement dans toutes les directions.
- 45 2. Fibre selon la revendication 1, dans laquelle ledit PEbdl a une densité dans la plage allant de 0,92 à 0,94 g/cm<sup>3</sup>.
- 50 3. Fibre selon la revendication 1 ou 2, dans laquelle ledit PEbdl a une teneur en comonomère alcène dans la plage allant de 3 à 20 % en poids du PEbdl.
4. Fibre selon l'une quelconque des revendications précédentes, dans laquelle la fibrille a un titre inférieur à 3,3 tex (30 deniers).
- 55 5. Fibre selon la revendication 4, dans laquelle la fibre a un titre dans la plage allant de 0,05 à 1,7 tex (0,5-15 deniers).

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6. Fibre selon la revendication 5, dans laquelle la fibre a un titre dans la plage allant de 0,1 à 0,6 tex (1-5 deniers).
- 5 7. Fibre selon l'une quelconque des revendications précédentes, dans laquelle le PEbdl a un indice de fluidité à chaud de 20 à 100 g/10 minutes.
8. Fibre selon la revendication 7, dans laquelle l'indice de fluidité à chaud est de  $50 \pm 20$  g/10 min.
- 10 9. Fibre selon l'une quelconque des revendications précédentes, dans laquelle le PEbdl a une densité de 0,92 à 0,93 g/cm<sup>3</sup>.
10. Fibre selon l'une quelconque des revendications précédentes, dans laquelle le comonomère alcène contient de 4 à 8 atomes de carbone.
- 15 11. Fibre selon l'une quelconque des revendications précédentes, dans laquelle le polyéthylène constitue de 25 à 35 % en poids du total.
12. Fibre selon la revendication 11, dans laquelle le polyéthylène constitue de 28 à 32 % en poids du total.
- 20 13. Fibre selon l'une quelconque des revendications précédentes, dans laquelle les fibrilles de polyéthylène ont une taille inférieure au micromètre et, pour la majeure partie d'entre elles, un diamètre inférieur à 0,05 micromètre.
- 25 14. Fibre selon l'une quelconque des revendications précédentes, dans laquelle ledit comonomère alcène est le 1-octène.
15. Fibre selon la revendication 14, dans laquelle le PEbdl a une densité d'environ 0,925 g/cm<sup>3</sup>, une teneur en octène de 5 à 10 % et un indice de fluidité à chaud de  $50 \pm 20$  g/10 min.
- 30 16. Procédé de fabrication de fibres à deux composants, comprenant l'expulsion, à travers une filière, d'un mélange intime de polypropylène (PP) fondu et de polyéthylène basse densité linéaire (PEbdl) fondu, caractérisé en ce que ledit PEbdl a un indice de fluidité à chaud [mesuré selon ASTM D-1238 (E)] dans la plage de 12 à 120 g/10 min et le rapport pondéral PP:PEbdl vaut de 80:20 à 55:45, ce par quoi les fibres comportent des fibrilles de PEbdl disposées de façon à s'étendre pratiquement dans toutes
- 35 les directions.
17. Procédé selon la revendication 16, dans lequel le mélangeage est effectué au moyen d'un appareil de mélangeage dynamique en trois dimensions.
- 40 18. Procédé selon la revendication 16 ou 17, dans lequel les composants du mélange sont ceux spécifiés dans l'une quelconque des revendications 2, 3, 7 à 12, 14 et 15 et/ou le mélange est filé pour donner des fibres telles que revendiquées dans l'une quelconque des revendications 4 à 6 et 13.

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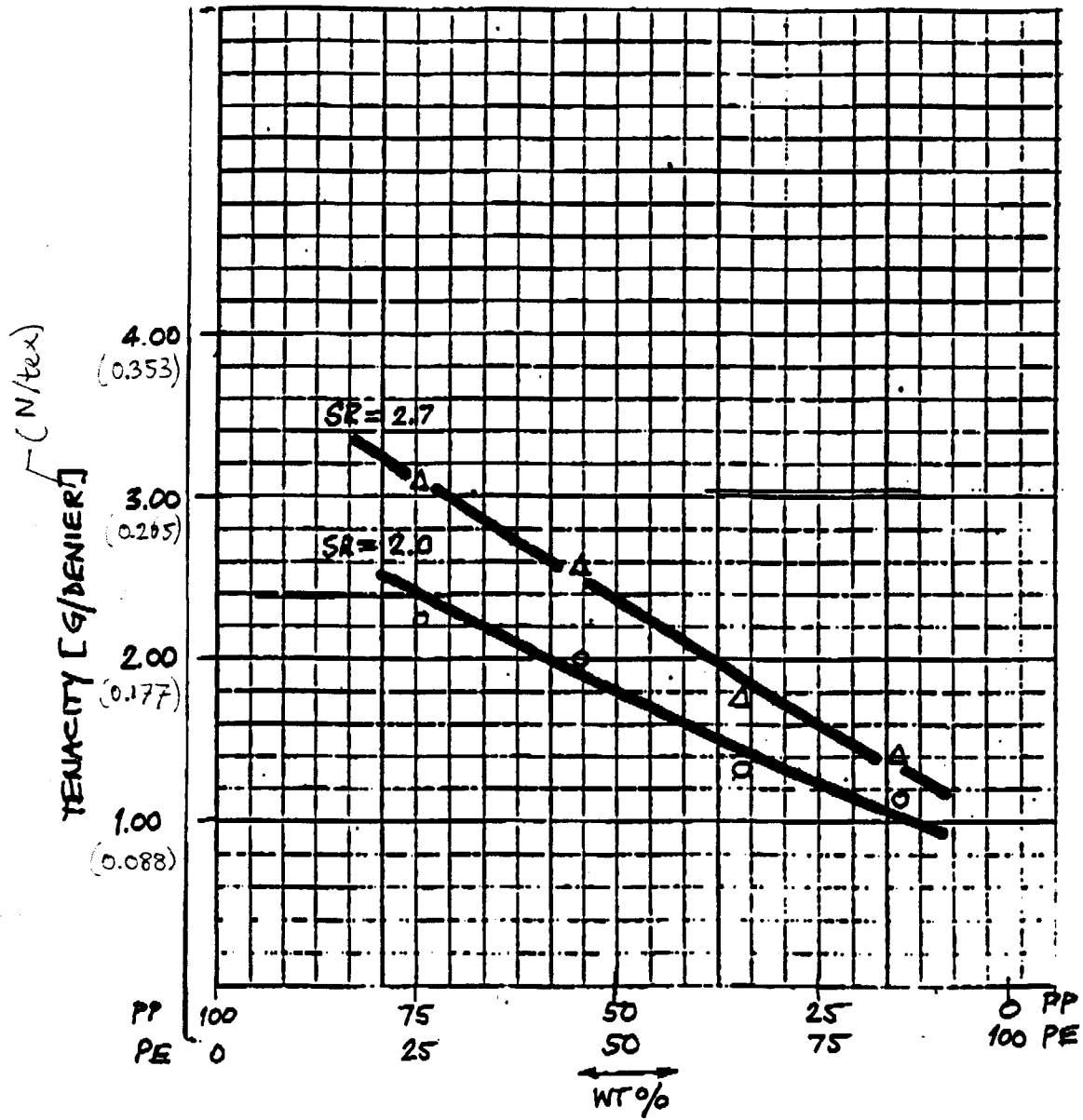


FIG. 1

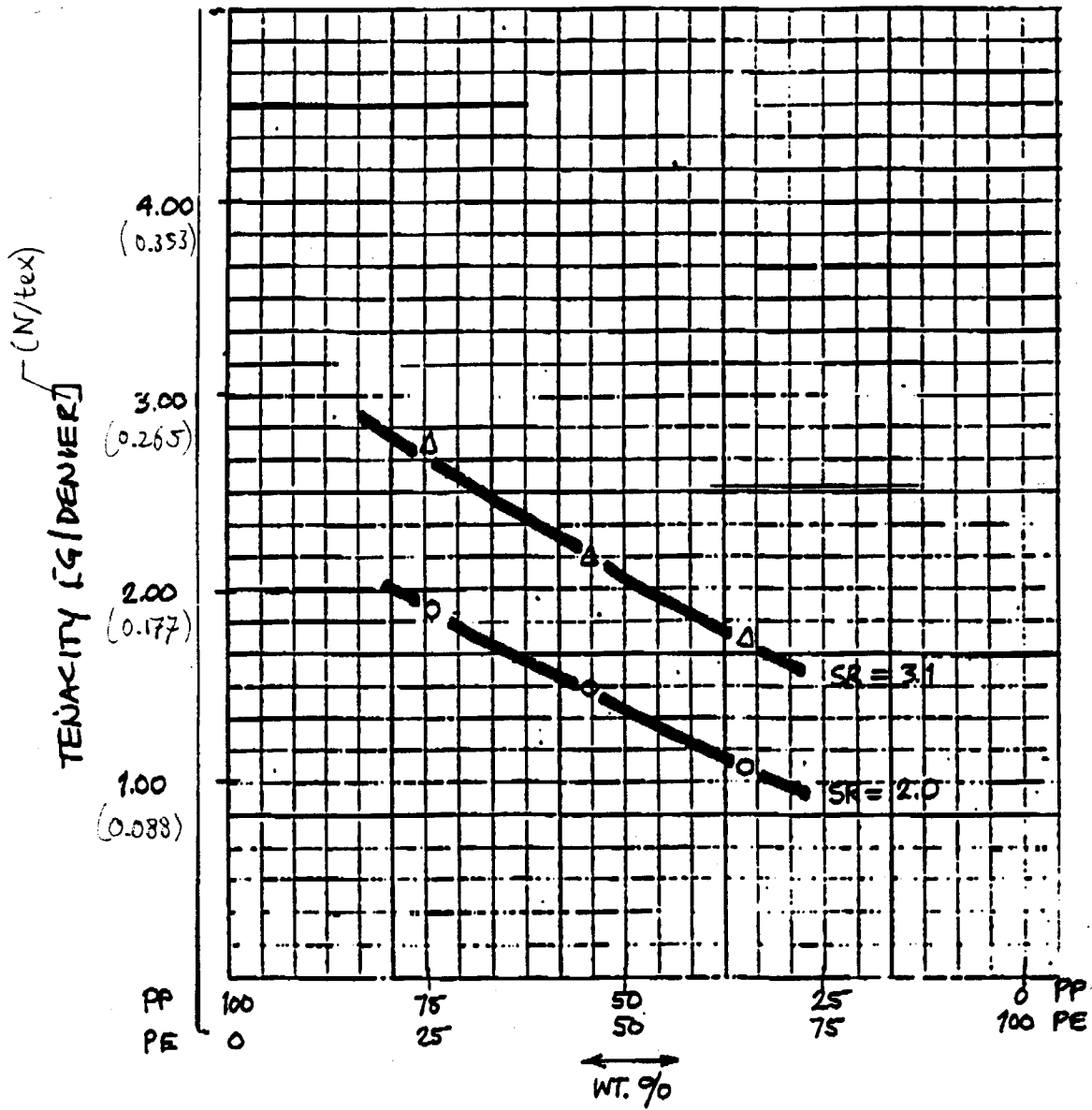


FIG. 2

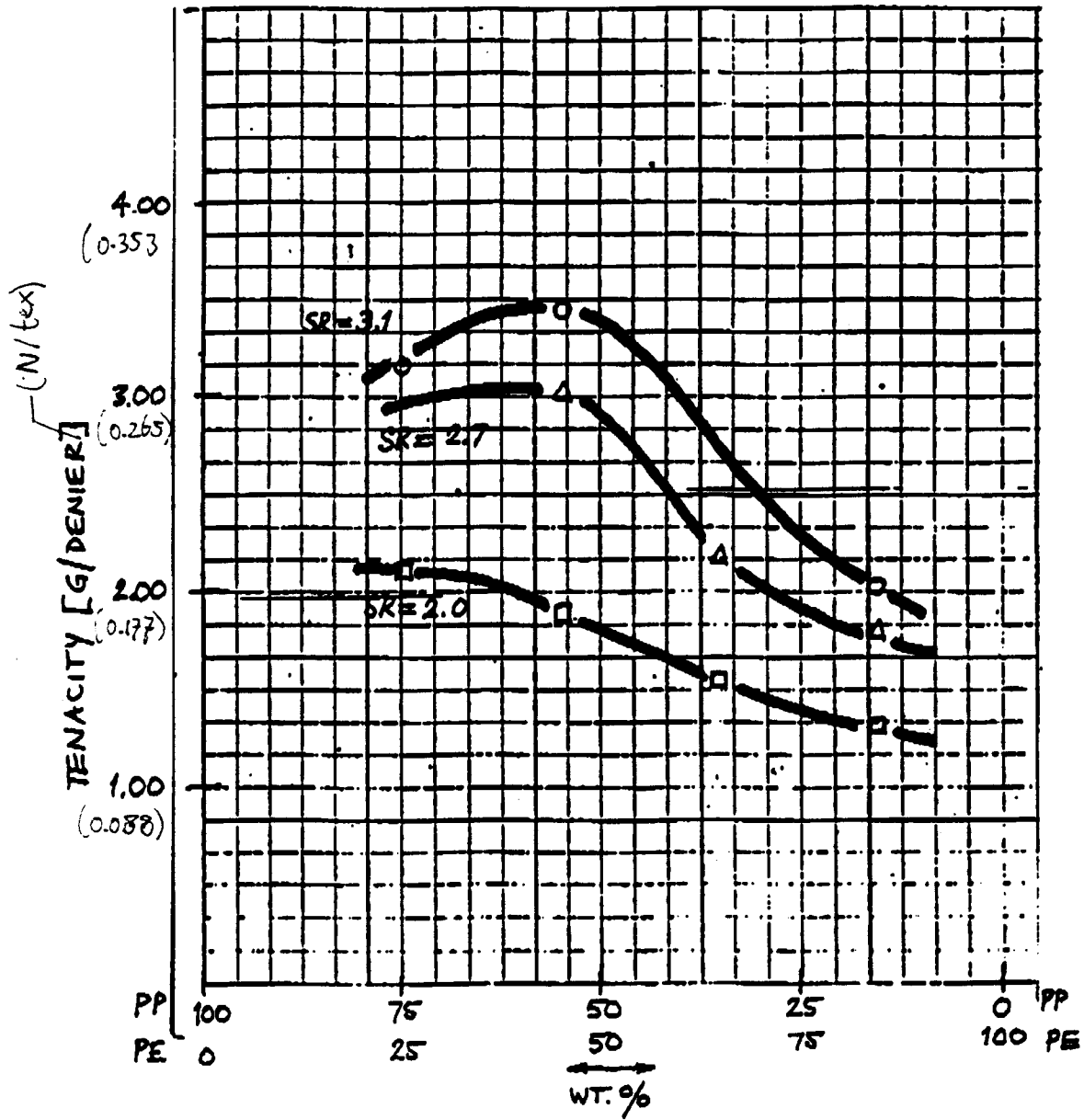


FIG. 3

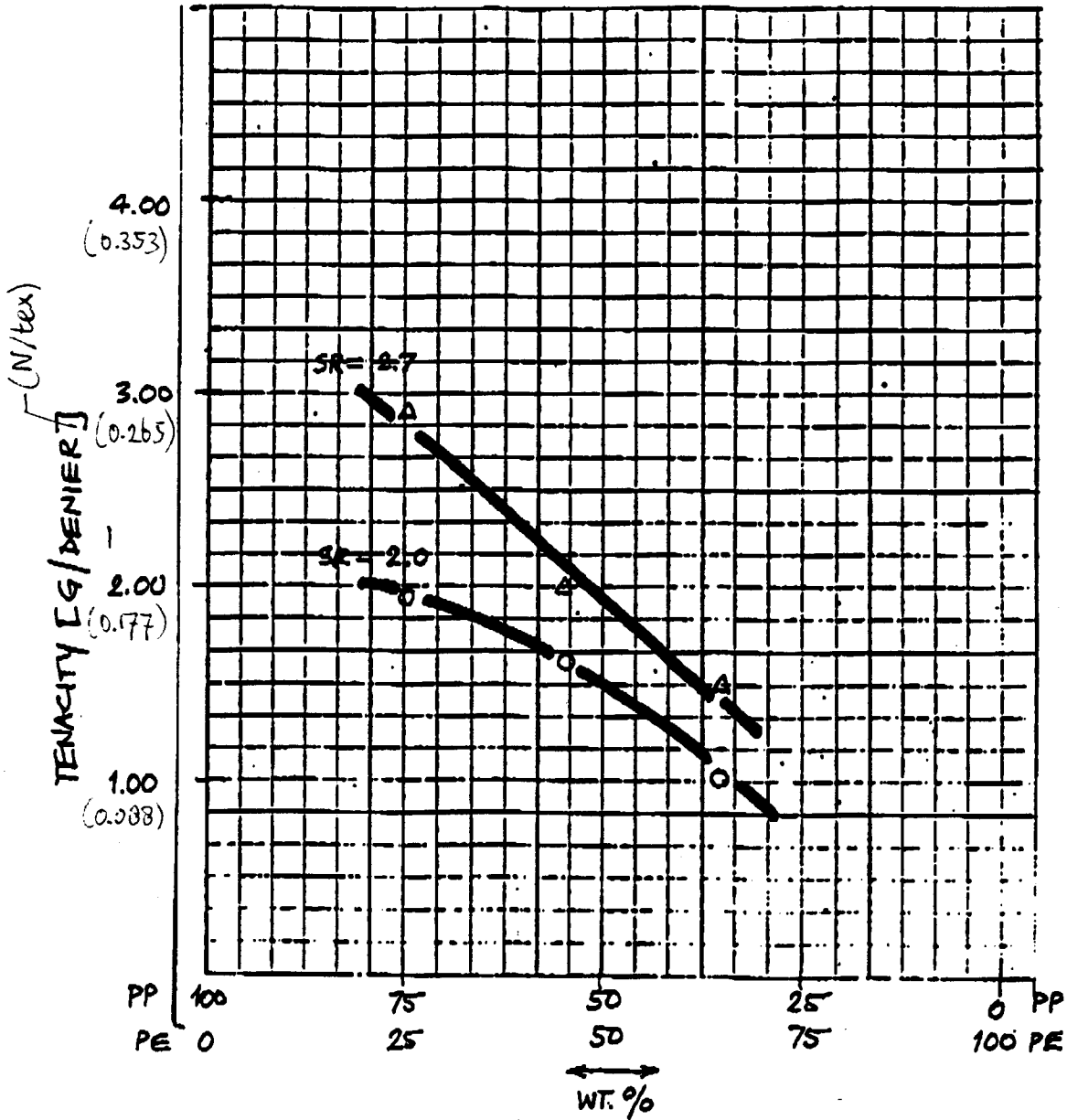


FIG. 4