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# (54) MULTIFILAMENT, METHOD FOR MANUFACTURING MULTIFILAMENT, STAPLE, AND METHOD FOR MANUFACTURING STAPLE

(57) The present invention is directed to a multifilament including a plurality of individual filaments. The individual filaments contain a poly(3-hydroxyalkanoate) resin and polycaprolactone, have a fineness of 1.0 to 5.0 dtex, and have a tensile strength of 2.5 cN/dtex or more.

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#### Description

#### **Technical Field**

<sup>5</sup> **[0001]** The present invention relates to a multifilament, a method for producing the multifilament, a staple, and a method for producing the staple.

#### **Background Art**

[0002] In recent years, waste plastics have become problematic since they have caused a significant impact on the global environment, for example, by affecting ecosystems, emitting hazardous gases during combustion, or generating a huge amount of combustion heat which contributes to global warming. As a solution to this problem, biodegradable plastics are under active development.

**[0003]** The biodegradable plastics include those obtained using plant-derived materials. Carbon dioxide emitted upon combustion of plant-derived biodegradable plastics is that which originally existed in the air, and does not cause an increase in the amount of atmospheric carbon dioxide. This is called "carbon neutrality". The carbon neutrality is emphasized under the Kyoto Protocol which specifies a carbon dioxide reduction goal, and the active use of carbon-neutral plastics is desired.

**[0004]** Nowadays, in terms of biodegradability and carbon neutrality, aliphatic polyester resins are attracting attention as biodegradable plastics microbially produced using plant-derived materials as carbon sources. Particular attention is being directed to polyhydroxyalkanoate resins.

**[0005]** Patent Literature 1 discloses a multifilament including a plurality of individual filaments containing a 3-hydroxyalkanoate polymer.

#### 25 Citation List

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#### **Patent Literature**

[0006] PTL 1: WO 2015/029316

#### **Summary of Invention**

#### **Technical Problem**

<sup>35</sup> **[0007]** A demand for a multifilament with increased strength can arise in the future.

[0008] However, sufficient investigations have not been made as to a multifilament having high strength.

[0009] A first object of the present invention is therefore to provide a multifilament having high strength.

[0010] A second object is to obtain a staple including a cut piece of the multifilament.

#### 40 Solution to Problem

[0011] A first aspect of the present invention relates to a multifilament including a plurality of individual filaments, wherein

the individual filaments contain a poly(3-hydroxyalkanoate) resin and polycaprolactone,

the individual filaments have a fineness of 1.0 to 5.0 dtex, and

the individual filaments have a tensile strength of 2.5 cN/dtex or more.

[0012] Preferably, a mass ratio of the poly(3-hydroxyalkanoate) resin to the polycaprolactone is from 30/70 to 80/20.

**[0013]** Preferably, the poly(3-hydroxyalkanoate) resin contains a structural unit represented by the following formula (1): [-CHR-CH<sub>2</sub>CO-O-] (1), wherein R is an alkyl group represented by  $C_pH_{2p+1}$  and p is an integer from 1 to 15.

[0014] Preferably, the individual filaments have a matrix-domain structure including a matrix and a domain,

the matrix contains the poly(3-hydroxyalkanoate) resin, and the domain contains the polycaprolactone.

[0015] A second aspect of the present invention relates to a staple including a cut piece of the multifilament, wherein

the staple has a crimped structure, and

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the staple has an average length of 200 mm or less.

**[0016]** A third aspect of the present invention relates to a multifilament production method for producing a multifilament including a plurality of individual filaments by melt spinning using a spinning nozzle having a plurality of discharge holes, the multifilament production method including the steps of:

- (A) discharging a melt from the plurality of discharge holes to obtain a plurality of raw filaments in a molten state;
- (B) blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments in the molten state to cool the plurality of raw filaments; and
- (C) stretching the plurality of cooled raw filaments by a stretching roll unit to obtain the multifilament, wherein

the melt contains a poly(3-hydroxyalkanoate) resin and polycaprolactone, and the individual filaments have a fineness of 1.0 to 5.0 dtex.

[0017] Preferably, the individual filaments have a tensile strength of 2.5 cN/dtex or more.

[0018] Preferably, in the step (C), the plurality of stretched raw filaments are heated by a heat treatment roll unit to obtain the multifilament.

[0019] Preferably, the raw filaments used in the step (C) have a fineness of 5.0 to 15.0 dtex.

**[0020]** Preferably, in the step (C), the plurality of raw filaments cooled in the step (B) are received on and heated by a receiving roll unit having a temperature that is 25°C or above but below 50°C, the plurality of raw filaments heated by the receiving roll unit are stretched by the stretching roll unit, and the plurality of raw filaments stretched by the stretching roll unit are heated by a heat treatment roll unit having a temperature of 40 to 100°C.

**[0021]** Preferably, in the step (B), the plurality of cooled raw filaments are wound on a raw filament-winding roll unit, and in the step (C), the plurality of raw filaments wound on the raw filament-winding roll unit are stretched by the stretching roll unit.

[0022] Preferably, in the multifilament production method, a spin draw process is used to produce the multifilament.

**[0023]** Preferably, in the step (C), the plurality of raw filaments stretched by the stretching roll unit are heated by a heat treatment roll unit having a temperature of 25 to 100°C.

[0024] A fourth aspect of the present invention relates to a staple production method including:

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using the multifilament production method to obtain the multifilament; and cutting the multifilament to obtain a staple, wherein the staple has a crimped structure, and the staple has an average length of 200 mm or less.

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#### **Advantageous Effects of Invention**

[0025] The present invention can provide a multifilament having high strength.

[0026] Additionally, the present invention can provide a staple including a cut piece of the multi filament.

#### **Brief Description of Drawings**

#### [0027]

FIG. 1 is a schematic diagram of an apparatus used in steps (A) and (B) in a first embodiment.

FIG. 2 is a schematic diagram of an apparatus used in step (C) in the first embodiment.

FIG. 3 is a schematic diagram of an apparatus used in a second embodiment.

### **Description of Embodiments**

«Multifilament»

[0028] A multifilament according to one embodiment will be described first.

[0029] The multifilament according to the present embodiment includes a plurality of individual filaments.

[0030] The individual filaments contain a poly(3-hydroxyalkanoate) resin and polycaprolactone (PCL).

**[0031]** The individual filaments have a fineness of 1.0 to 5.0 dtex.

[0032] The individual filaments have a tensile strength of 2.5 cN/dtex or more.

[0033] The individual filaments are obtained by forming a polymer composition containing a polymer component into

filaments.

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[0034] The polymer composition may further contain an additive.

[0035] The polymer component contains a poly(3-hydroxyalkanoate) resin and polycaprolactone.

**[0036]** The polymer component may further contain an additional polymer as well as the poly(3-hydroxyalkanoate) resin and polycaprolactone.

[0037] The poly(3-hydroxyalkanoate) resin is a polyester whose monomer is a 3-hydroxyalkanoate.

[0038] The poly(3-hydroxyalkanoate) resin is a polymer having biodegradability.

**[0039]** As used in the present embodiment, the term "biodegradability" refers to the property of being degradable into low-molecular-weight compounds by microorganisms in nature. Specifically, the possession or lack of biodegradability can be determined based on any of various tests each of which is adapted for a different environment. Examples of tests adapted for aerobic conditions include ISO 14855 (compost) and ISO 14851 (activated sludge), and examples of tests adapted for anaerobic conditions include ISO 14853 (aqueous phase) and ISO 15985 (solid phase). The microbial degradability in seawater can be evaluated by measurement of biochemical oxygen demand.

[0040] The poly(3-hydroxyalkanoate) resin may be a homopolymer or a copolymer.

15 **[0041]** The poly(3-hydroxyalkanoate) resin preferably contains a structural unit represented by the following formula (1).

[-CHR-CH<sub>2</sub>-CO-O-] (1

[0042] In the formula (1), R is an alkyl group represented by  $C_pH_{2p+1}$  and p is an integer from 1 to 15.

[0043] The poly(3-hydroxyalkanoate) resin preferably contains 3-hydroxybutyrate as a structural unit.

**[0044]** Examples of the poly(3-hydroxyalkanoate) resin containing 3-hydroxybutyrate as a structural unit include P3HB, P3HB3HV, P3HB3HV, P3HB4HB,

poly(3-hydroxybutyrate-co-3-hydroxyoctanoate), and poly(3-hydroxybutyrate-co-3-hydroxyoctadecanoate).

P3HB refers to poly(3-hydroxybutyrate).

P3HB3HH refers to poly(3-hydroxybutyrate-co-3-hydroxyhexanoate).

P3HB3HV refers to poly(3-hydroxybutyrate-co-3-hydroxyvalerate).

P3HB4HB refers to poly(3-hydroxybutyrate-co-4-hydroxybutyrate).

**[0045]** The poly(3-hydroxyalkanoate) resin preferably includes P3HB because P3HB has the function of accelerating its own crystallization and crystallization of another poly(3-hydroxyalkanoate) resin.

**[0046]** In terms of ensuring both high biodegradability and high moldability, the poly(3-hydroxyalkanoate) resin is preferably, but not limited to, P3HB, P3HB3HV, or P3HB4HB.

**[0047]** In terms of increasing the strength of the multifilament according to the present embodiment and increasing the moldability, the poly(3-hydroxyalkanoate) resin is preferably P3HB3HH.

**[0048]** The poly(3-hydroxyalkanoate) resin preferably contains 85.0 to 99.5 mol%, more preferably 85.0 to 97.0 mol%, 3-hydroxybutyrate as a structural unit.

**[0049]** When the poly(3-hydroxyalkanoate) resin contains 85.0 mol% or more 3-hydroxybutyrate as a structural unit, the multifilament according to the present embodiment has high stiffness.

**[0050]** When the poly(3-hydroxyalkanoate) resin contains 99.5 mol% or less 3-hydroxybutyrate as a structural unit, the multifilament according to the present embodiment has high flexibility.

**[0051]** The polymer component may contain only one poly(3-hydroxyalkanoate) resin as described above or two or more such poly(3-hydroxyalkanoate) resins.

**[0052]** In the case where the poly(3-hydroxyalkanoate) resin includes a copolymer (such as P3HB3HH), the poly(3-hydroxyalkanoate) resin may include two or more copolymers differing in the average proportions of structural units.

**[0053]** The weight-average molecular weight of the poly(3-hydroxyalkanoate) resin is preferably from 50,000 to 3,000,000 and more preferably from 100,000 to 1,500,000.

**[0054]** When the weight-average molecular weight of the poly(3-hydroxyalkanoate) resin is 3,000,000 or less, the multifilament according to the present embodiment can be easily formed.

**[0055]** When the weight-average molecular weight of the poly(3-hydroxyalkanoate) resin is 50,000 or more, the strength of the multifilament according to the present embodiment can be increased.

**[0056]** The weight-average molecular weight in the present embodiment refers to that determined from a polystyrene-equivalent molecular weight distribution measured by gel permeation chromatography (GPC) using a chloroform eluent. The column used in the GPC may be any column suitable for measurement of the molecular weight.

[0057] The polycaprolactone is a polymer resulting from ring-opening polymerization of  $\epsilon$ -polycaprolactone.

[0058] The polycaprolactone has biodegradability like the poly(3-hydroxyalkanoate) resin.

[0059] Additionally, the fact that the individual filaments contain the polycaprolactone leads to high strength of the multifilament according to the present embodiment.

**[0060]** The weight-average molecular weight of the polycaprolactone is preferably from 5,000 to 500,000 and more preferably from 10,000 to 200,000.

**[0061]** When the weight-average molecular weight of the polycaprolactone is 500,000 or less, the multifilament according to the present embodiment can be easily formed.

**[0062]** When the weight-average molecular weight of the polycaprolactone is 5,000 or more, the strength of the multifilament according to the present embodiment can be increased.

[0063] The additional polymer preferably has biodegradability.

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[0064] Examples of the additional polymer having biodegradability include polylactic acid, polybutylene succinate, polybutylene succinate adipate, polybutylene adipate terephthalate, polyethylene succinate, polyvinyl alcohol, polyglycolic acid, native starch, modified starch, cellulose acetate, chitosan, and poly(4-hydroxyalkanoate) resins.

[0065] The polymer composition may contain one such additional polymer or two or more such additional polymers.

**[0066]** The total amount of the poly(3-hydroxyalkanoate) resin and the polycaprolactone contained in the polymer component is preferably 50 mass% or more, more preferably 80 mass% or more, and even more preferably 90 mass% or more.

**[0067]** The mass ratio of the poly(3-hydroxyalkanoate) resin to the polycaprolactone is preferably from 30/70 to 80/20, more preferably from 40/60 to 75/25, and even more preferably from 55/45 to 70/30.

**[0068]** When the mass ratio of the poly(3-hydroxyalkanoate) resin to the polycaprolactone is 30/70 or more, the advantage is that the multifilament according to the present embodiment has high heat resistance.

**[0069]** When the mass ratio of the poly(3-hydroxyalkanoate) resin to the polycaprolactone is 80/20 or less, the advantage is that the multifilament according to the present embodiment is likely to have high strength.

**[0070]** Preferably, the individual filaments have a matrix-domain structure including a matrix and a domain, the matrix contains the poly(3-hydroxyalkanoate) resin, and the domain contains the polycaprolactone.

**[0071]** The multifilament according to the present embodiment has the advantage of being able to exhibit high heat resistance when the individual filaments have the above matrix-domain structure.

[0072] A possible reason for this advantage of the multifilament according to the present embodiment is as follows.

**[0073]** The matrix-domain structure is also called a sea-island structure. In the matrix-domain structure, the matrix is a continuous phase. In contrast, the domain is a discontinuous phase made up of discrete regions which are like islands in the sea.

[0074] Thus, mainly the properties of the matrix emerge as the properties of the individual filaments.

[0075] The poly(3-hydroxyalkanoate) resin has higher heat resistance than the polycaprolactone.

**[0076]** This is presumably why the multifilament according to the present embodiment has high heat resistance when the individual filaments have the structure as described above.

**[0077]** By virtue of containing a biodegradable polymer, the multifilament according to the present embodiment can be easily degraded in the environment when discarded into the environment. Thus, the use of this multifilament leads to a reduced impact on the environment.

**[0078]** Examples of the additive include a nucleating agent, a lubricant, a stabilizer (such as an antioxidant or ultraviolet absorber), a colorant (such as a dye or pigment), a plasticizer, an inorganic filler, an organic filler, and an antistat.

**[0079]** The polymer composition preferably contains a nucleating agent in order to accelerate the crystallization of the poly(3-hydroxyalkanoate) resin.

**[0080]** The nucleating agent is a compound that has an accelerating effect on the crystallization of the poly(3-hydroxyalkanoate) resin. The nucleating agent has a higher melting point than the poly(3-hydroxyalkanoate) resin.

[0081] Examples of the nucleating agent include: inorganic substances (such as boron nitride, titanium oxide, talc, layered silicates, calcium carbonate, sodium chloride, and metal phosphates); sugar alcohol compounds derived from natural sources (such as pentaerythritol, erythritol, galactitol, mannitol, and arabitol); polyvinyl alcohol; chitin; chitosan; polyethylene oxide; salts of aliphatic carboxylic acids; aliphatic alcohols; esters of aliphatic carboxylic acids; dicarboxylic acid derivatives (such as dimethyl adipate, dibutyl adipate, diisodecyl adipate, and dibutyl sebacate); cyclic compounds having in the molecule C=O and a functional group selected from NH, S, and O (such as indigo, quinacridone, and quinacridone magenta); sorbitol derivatives (such as bis(benzylidene) sorbitol and bis(p-methylbenzylidene) sorbitol); compounds (such as pyridine, triazine, and imidazole) which have a nitrogen-containing heteroaromatic core (such as a pyridine ring, a triazine ring, or an imidazole ring); phosphoric ester compounds; bisamides of higher fatty acids; metal salts of higher fatty acids; and branched polylactic acid.

[0082] P3HB, which is an example of the poly(3-hydroxyalkanoate) resin, can be used also as the nucleating agent.

[0083] One of the above-mentioned substances may be used alone, or two or more thereof may be used in combination.

**[0084]** The sugar alcohol compounds, polyvinyl alcohol, chitin, and chitosan are preferred as the nucleating agent in terms of increasing the rate of crystallization of the poly(3-hydroxyalkanoate) resin and in terms of the compatibility with and affinity for the poly(3-hydroxyalkanoate) resin.

[0085] Among the sugar alcohol compounds, pentaerythritol is preferred.

**[0086]** The amount of the nucleating agent contained in the polymer composition is preferably 0.05 parts by mass or more, more preferably 0.1 parts by mass or more, and even more preferably 0.5 parts by mass or more per 100 parts by mass of the poly(3-hydroxyalkanoate) resin. When the amount of the nucleating agent contained in the polymer composition is 0.05 parts by mass or more per 100 parts by mass of the poly(3-hydroxyalkanoate) resin, the advantage is that the crystallization of the poly(3-hydroxyalkanoate) resin can be further accelerated.

**[0087]** The amount of the nucleating agent contained in the polymer composition is preferably 10 parts by mass or less, more preferably 8 parts by mass or less, and even more preferably 5 parts by mass or less per 100 parts by mass of the poly(3-hydroxyalkanoate) resin. When the amount of the nucleating agent contained in the polymer composition is 10 parts by mass or less per 100 parts by mass of the poly(3-hydroxyalkanoate) resin, the advantage is that during making of the multifilament from a melt of the polymer composition, the viscosity of the melt can be made low enough to easily accomplish the making of the multifilament.

**[0088]** P3HB can be used as the poly(3-hydroxyalkanoate) resin and can function as the nucleating agent. Thus, in the case where the polymer composition contains P3HB, the amount of P3HB is included both in the amount of the poly(3-hydroxyalkanoate) resin and in the amount of the nucleating agent.

**[0089]** The polymer composition preferably contains the lubricant. When the individual filaments contain the lubricant, the lubricity of the individual filaments is enhanced, and fusion between the individual filaments can be prevented.

[0090] Examples of the lubricant include a compound having an amide bond.

**[0091]** The compound having an amide bond preferably includes at least one selected from lauramide, myristamide, stearamide, behenamide, and erucamide.

**[0092]** The amount of the lubricant contained in the polymer composition is preferably 0.05 parts by mass or more, more preferably 0.1 parts by mass or more, and even more preferably 0.5 parts by mass or more per 100 parts by mass of the polymer component. When the amount of the lubricant contained in the polymer composition is 0.05 parts by mass or more per 100 parts by mass of the polymer component, the advantage is that the individual filaments have high lubricity.

**[0093]** The amount of the lubricant contained in the polymer composition is preferably 12 parts by mass or less, more preferably 10 parts by mass or less, even more preferably 8 parts by mass or less, and most preferably 5 parts by mass or less per 100 parts by mass of the polymer component. When the amount of the lubricant contained in the polymer composition is 12 parts by mass or less per 100 parts by mass of the polymer component, the advantage is that bleed-out of the lubricant to the surface of the multifilament can be prevented.

[0094] The individual filaments have a fineness of 1.0 to 5.0 dtex.

[0095] The fineness of the individual filaments is preferably more than 1.0 dtex and more preferably 1.5 dtex or more.

[0096] The fineness of the individual filaments is preferably 4.7 dtex or less and more preferably 4.5 dtex or less.

**[0097]** In the present embodiment, the fineness of the individual filaments refers to the thickness of the filaments and is defined as the mass per unit length. The mass (g) per 10,000 m is expressed in units of dtex. Specifically, the fineness is measured by an automatic vibroscope method.

**[0098]** The individual filaments have a tensile strength of 2.5 cN/dtex or more. The tensile strength of the individual filaments is preferably 2.6 cN/dtex or more. Although it is more preferable for the individual filaments to have a higher tensile strength, the tensile strength of the individual filament is, for example, 10 cN/dtex or less.

**[0099]** The tensile strength of the individual filaments is not limited to a particular range insofar as the flexibility and toughness are not below the levels required for the intended purpose. The tensile strength of the individual filaments may be 10 cN/dtex or less.

**[0100]** The tensile strength of the individual filaments can be measured at an initial length of 20 mm and a speed of 20 mm/min based on JIS L 1015:2010 "Test methods for man-made staple fibers".

**[0101]** The multifilament according to the present embodiment may be used by itself. Alternatively, a plurality of such multifilaments may be used in the form of a braid or sheet.

**[0102]** The multifilament according to the present embodiment can be used in various environments such as marine and land environments.

**[0103]** The multifilament according to the present embodiment can be used, for example, in a fishing net, a marine rope, a net for aquaculture, a fishing line, a net for agriculture, artificial turf, a net for sandbags, or a waterproof sheet. The net for sandbags can be used, for example, for bank protection work.

<<Staple>>

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**[0104]** A staple according to the present embodiment is a staple including a cut piece of the multifilament according to the present embodiment.

**[0105]** The staple according to the present embodiment has a crimped structure. In other words, the staple according to the present embodiment is a crimped yarn.

[0106] The staple according to the present embodiment has an average length of 200 mm or less. The average length

is not limited to a particular range and may be in any range up to 200 mm. The average length may be set as appropriate depending on the intended purpose of the staple.

**[0107]** In terms of the usability for various purposes, the average length of the staple according to the present embodiment is preferably 160 mm or less and in particular from 1.0 to 100 mm.

[0108] The average length of the staple refers to "average value of fiber lengths" as determined by "c) Method C (replacement method)" as specified in JIS L 1015:2021 "Test methods for man-made staple fibers", "8.4 Fiber length", "8.4.1 Average fiber length".

[0109] The staple according to the present embodiment can be used in the form of a dry non-woven fabric.

**[0110]** The dry non-woven fabric can be used, for example, as a hygiene material, a medical material, a daily life material, a waterproof material, or an interior material for automobiles. Specifically, the dry non-woven fabric can be used, for example, as a diaper, a filter, a towel, a wipe, a gauze, a napkin, or a carpet.

<<Multifilament Production Method>>

15 [0111] A multifilament production method according to the present embodiment is a method for producing a multifilament including a plurality of individual filaments by melt spinning using a spinning nozzle having a plurality of discharge holes.

**[0112]** The multifilament production method according to the present embodiment includes the steps of: (A) discharging a melt from the plurality of discharge holes to obtain a plurality of raw filaments in a molten state; (B) blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments in the molten state to cool the plurality of raw filaments; and (C) stretching the plurality of cooled raw filaments by a stretching roll unit to obtain the multifilament.

**[0113]** The melt contains a poly(3-hydroxyalkanoate) resin and polycaprolactone.

[0114] The individual filaments have a fineness of 1.0 to 5.0 dtex.

[0115] Preferably, the individual filaments have a tensile strength of 2.5 cN/dtex or more.

**[0116]** In the step (A), the melt is the above-described polymer composition in a molten state.

<First Embodiment: Post-stretching Process (Sequential Stretching Process)>

**[0117]** Hereinafter, a multifilament production method according to a first embodiment will be described with reference to FIGS. 1 and 2. The example described below is one in which a multifilament is produced by a post-stretching process (also referred to as a "sequential stretching process").

**[0118]** In the multifilament production method according to the first embodiment, the plurality of raw filaments cooled by the gas are wound on a raw filament-winding roll unit, and the plurality of raw filaments wound on the raw filament-winding roll unit are stretched by the stretching roll unit.

(Step (A))

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[0119] In the step (A), as shown in FIG. 1, materials of the melt are first placed into a material feeder 101.

**[0120]** Next, the materials fed from the material feeder 101 are kneaded under heating by a kneading extruder 102 to obtain the melt.

**[0121]** The kneading extruder 102 is a screw extruder. The kneading extruder 102 may be a single-screw extruder or a twin-screw extruder.

**[0122]** Subsequently, a spinning nozzle 104 having a plurality of discharge holes is used; specifically, the melt obtained in the kneading extruder 102 is discharged from the plurality of discharge holes to obtain a plurality of raw filaments 100A in a molten state.

**[0123]** The flow rate of the melt discharged from the plurality of discharge holes of the spinning nozzle 104 is adjusted by a gear pump 103.

[0124] The temperature of the spinning nozzle 104 is, for example, from 160 to 180°C.

**[0125]** The discharge holes are not limited to a particular shape, size, or number. As to the size of the discharge holes, for example, when the discharge holes are circular, the diameter of the discharge holes is preferably from 0.1 to 3.0 mm. The number of the discharge holes, although depending on the size of the discharge holes, may be, for example, 15 or more and 1000 or less.

**[0126]** The spinning nozzle flow speed, i.e., the speed at which the melt is discharged from the spinning nozzle 104, is preferably from 0.05 to 20 m/min, more preferably from 1.0 to 10 m/min, and even more preferably from 0.5 to 5.0 m/min.

**[0127]** The discharge amount by which the melt is discharged from the discharge holes of the spinning nozzle 104 is preferably 0.10 g/min/hole or more and more preferably 0.15 g/min/hole or more. The discharge amount is preferably less than 1.0 g/min/hole and more preferably 0.90 g/min/hole or less.

(Step (B))

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[0128] In the step (B), a gas having a temperature of 3 to 15°C is blown onto the plurality of raw filaments 100A obtained in a molten state in the step (A), and thus the plurality of raw filaments 100A are cooled.

**[0129]** In the step (B), cooling the raw filaments 100A by a gas having a temperature of 3 to 15°C can shorten the time during which the temperature of the raw filaments 100A is in a temperature range where the polycaprolactone and poly(3-hydroxyalkanoate) resin constituting the raw filaments 100A crystallize, and thus can retard the crystallization of the polycaprolactone and poly(3-hydroxyalkanoate) resin. This can reduce the degree of hardening of the raw filaments 100A. Thus, stretching of the raw filaments 100A in the step (C) can be easily performed. As such, the strength of the multifilament can be easily increased.

**[0130]** In the first embodiment, the raw filaments 100A are cooled by a gas having a temperature of 3 to 15°C in a first cooling box 105, and the raw filaments 100A cooled in the first cooling box 105 are further cooled by a gas having a temperature of 3 to 15°C in a second cooling box 106.

**[0131]** The temperature of a gas blown onto the plurality of raw filaments 100A obtained in a molten state in the step (A) is from 3 to 15°C and preferably from 3.0 to 6.0°C.

**[0132]** The "temperature of a gas blown onto the plurality of raw filaments 100A obtained in a molten state in the step (A)" refers to the temperature that the gas has when contacting the raw filaments 100A.

**[0133]** The speed of the gas blown onto the plurality of raw filaments 100A obtained in a molten state in the step (A) is not limited to a particular range, but is preferably from 0.1 to 5 m/s and more preferably from 0.1 to 3 m/s.

[0134] When the speed of the gas is 0.1 m/s or more, the cooling effect of the gas can be easily achieved.

**[0135]** When the speed of the gas is 5 m/s or less, the raw filaments 100A discharged in a molten state from the spinning nozzle 104 are not much shaken by the gas. This reduces the occurrence of fusion between the raw filaments 100A in a molten state and/or the occurrence of filament breakage and therefore ensures high spinning stability.

**[0136]** The "speed of the gas blown onto the plurality of raw filaments 100A obtained in a molten state in the step (A)" refers to the speed that the gas has when contacting the raw filaments 100A.

[0137] Examples of the gas include air and inert gases (such as nitrogen gas and argon gas).

**[0138]** In the step (B), the raw filaments 100A cooled by the gas having a temperature of 3 to 15°C are received on a first receiving roll unit 107. The first receiving roll unit 107 includes two rolls. The first receiving roll unit 107 may include one roll or three or more rolls.

[0139] In the step (B), the plurality of raw filaments 100 A received on the first receiving roll unit 107 are wound on a raw filament-winding roll unit 112.

**[0140]** In the first embodiment, the plurality of raw filaments 100A received on the first receiving roll unit 107 are transferred to the raw filament-winding roll unit 112 by using a first transfer roll unit 108, a second transfer roll unit 109, a third transfer roll unit 110, and a fourth transfer roll unit 111.

[0141] Each of the transfer roll units includes two rolls in FIG. 1. Each of the transfer roll units may include one roll or three or more rolls.

**[0142]** In the step (B), the plurality of raw filaments 1 00A are preferably cooled to 50°C or below and more preferably cooled to 40°C or below. In the step (B), the temperature to which the plurality of raw filaments 100A are cooled is, for example, 0°C or above and in particular 10°C or above.

[0143] In the step (B), the plurality of raw filaments may be cooled to 50°C or below by blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments. In the step (B), the plurality of raw filaments 100A may be cooled to a certain extent by blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments 100A and then the plurality of raw filaments 100A may be cooled to 50°C or below by ambient air during transfer of the raw filaments 100A from the first receiving roll unit 107 to the raw filament-winding roll unit 112.

<sup>5</sup> **[0144]** In view of stretching of the raw filaments 100A in the step (C), it is preferable that the raw filaments 100A be little or not stretched in the step (B).

**[0145]** That is, the stretching ratio in the step (B) is preferably 1.5 or less, more preferably 1.2 or less, even more preferably 1.1 or less, and most preferably 1.0. The stretching ratio in the step (B) is 1.0 or more.

[0146] The stretching ratio in the step (B) can be determined by the following equation.

50 **[0147]** Stretching ratio in step (B) = speed (m/min) of raw filament-winding roll unit/speed (m/min) of receiving roll unit used in step (B) ("first receiving roll unit 107" in the case of the first embodiment)

**[0148]** The speed (m/min) of the raw filament-winding roll unit refers to the length by which the raw filaments are wound on the raw filament-winding roll unit per unit time.

**[0149]** The speed (m/min) of the receiving roll unit used in the step (B) refers to the length by which the raw filaments are received on the receiving roll unit used in the step (B) ("first receiving roll unit 107" in the case of the first embodiment) per unit time.

**[0150]** In the first embodiment, if the calculated value of the stretching ratio in the step (B) is less than 1.0, the actual stretching ratio is assumed to be 1.0.

(Step (C))

[0151] In the step (C), as shown in FIG. 2, the plurality of raw filaments 100A cooled to 50°C or below in the step (B) are heated, and the plurality of heated raw filaments 100A are stretched by a stretching roll unit 114.

**[0152]** In the step (C), stretching the plurality of raw filaments 100A can increase the degree of orientation of the polymer component contained in the raw filaments and thus can increase the tensile strength of the raw filaments. This can result in an increased strength of the multifilament.

**[0153]** To increase the degree of orientation of the polymer component, it is desired to stretch the raw filaments in a temperature range suitable for increasing the degree of orientation of the polymer component. This is because, if the raw filaments are stretched at a temperature above the temperature range, the polymer component is brought into a molten state and thus the stretching does not increase the degree of orientation of the polymer component very much. Another reason is that if the raw filaments are stretched at a temperature below the temperature range, the polymer component becomes so hard as to make stretching of the raw filaments difficult and that in this case, forcibly pulling the raw filaments to stretch the raw filaments could cause breakage of the raw filaments, leading to a failure to produce the multifilament

**[0154]** In the first embodiment, the plurality of raw filaments 100A cooled to 50°C or below in the step (B) are heated, and the plurality of heated raw filaments 100A are stretched by the stretching roll unit 114. Thus, the temperature of the plurality of raw filaments to be stretched is easier to control within a temperature range suitable for increasing the degree of orientation of the polymer component than in the case of a spin draw process in which a plurality of raw filaments are stretched while being cooled by ambient air. As such, the degree of orientation of the polymer component of the plurality of raw filaments can be easily increased.

[0155] Thus, in the first embodiment, the strength of the multifilament can be easily increased.

[0156] The fineness of the raw filaments used in the step (C) is preferably from 5.0 to 15.0 dtex and more preferably from 6.0 to 10 dtex.

**[0157]** For the fineness of the raw filaments used in the step (C), a relationship expressed by the following equation is approximately established.

Fineness (dtex) of raw filaments used in step (C) =  $(((a \times 1000/60)/b \times 10000)/c)/d$ 

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- a: Amount (kg/h) of melt discharged from spinning nozzle 104
- b: Speed (m/min) of receiving roll unit used in step (B) ("first receiving roll unit 107" in the case of the first embodiment)
- c: Number of discharge holes of spinning nozzle 104
- d: Stretching ratio (-) in step (B)

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**[0158]** Thus, the fineness of the raw filaments used in the step (C) can be controlled by adjusting the parameters such as b.

[0159] In the step (C), the plurality of raw filaments wound on the raw filament-winding roll unit 112 are received on a second receiving roll unit 113.

**[0160]** Subsequently, in the step (C), the raw filaments 100A received on the second receiving roll unit 113 are stretched by the stretching roll unit 114.

[0161] In the step (C), the multifilament obtained by the stretching is wound on a multifilament-winding roll unit 116.

[0162] In the step (C), the raw filaments 100A stretched by the stretching roll unit 114 may be transferred by a take-off roll unit 115.

**[0163]** The second receiving roll unit 113 includes two rolls. The second receiving roll unit 113 may include one roll or three or more rolls.

[0164] In the step (C), the plurality of raw filaments 1 00A are preferably heated by the second receiving roll unit 113.

[0165] In the step (C), heating the plurality of raw filaments 100A by the second receiving roll unit 113 makes it easy

to control the temperature of the raw filaments 100A within a temperature range suitable for increasing the degree of orientation of the polymer component contained in the plurality of raw filaments 100A, thus making it easy to increase the degree of orientation of the polymer component of the plurality of raw filaments 100A.

**[0166]** The temperature of the second receiving roll unit 113 is preferably 25°C or above but below 50°C and more preferably from 30 to 45°C.

**[0167]** When the temperature of the environment where the step (C) is performed is 25°C or above, the plurality of raw filaments 100A need not be heated by the second receiving roll unit 113.

**[0168]** The stretching roll unit 114 includes two rolls. The stretching roll unit 114 may include one roll or three or more rolls.

[0169] In the step (C), the plurality of raw filaments 100A may or may not be heated by the stretching roll unit 114.

That is, in the first embodiment, the stretching roll unit 114 may serve as a heat treatment roll unit.

**[0170]** The step (C) may employ the technique of heating the plurality of raw filaments 100A by the stretching roll unit 114 to accelerate the crystallization of the polymer component contained in the plurality of raw filaments 100A.

[0171] The temperature of the stretching roll unit (heat treatment roll unit) 114 is preferably from 40 to 100°C and more preferably from 50 to 90°C.

[0172] In the first embodiment, it is preferable for the take-off roll unit 115 to serve as a heat treatment roll unit.

**[0173]** The take-off roll unit 115 (heat treatment roll unit 115) includes two rolls. The take-off roll unit 115 (heat treatment roll unit 115) may include one roll or three or more rolls.

**[0174]** That is, in the step (C), the plurality of stretched raw filaments 100A are heated by the heat treatment roll unit 115 to obtain the multifilament.

**[0175]** In the step (C), the crystallization of the polymer component contained in the plurality of raw filaments 1 00A can be accelerated by heating the plurality of raw filaments 100A by the heat treatment roll unit 115.

**[0176]** The temperature of the take-off roll unit 115 (heat treatment roll unit 115) is preferably from 40 to 100°C and more preferably from 50 to 90°C.

[0177] Either or both the stretching roll unit 114 and take-off roll unit 115 may serve as heat treatment roll units.

[0178] The stretching ratio in the step (C) is preferably 2.0 or more. The stretching ratio in the step (C) is, for example, 10.0 or less.

**[0179]** When the stretching ratio in the step (C) is 2.0 or more, the degree of orientation of the polymer component of the plurality of raw filaments 100A can be further increased.

**[0180]** The stretching ratio in the step (C) can be determined by the following equation.

**[0181]** Stretching ratio in step (C) = speed (m/min) of multifilament-winding roll unit/speed (m/min) of receiving roll unit used in step (C) ("second receiving roll unit 113" in the case of the first embodiment)

[0182] In the step (C), the relaxation rate determined by the following equation is preferably from 5 to 15%.

Relaxation rate (%) = ((speed of stretching roll unit 114 – speed of

multifilament-winding roll unit 116)/speed of multifilament-winding roll unit 116) × 100

**[0183]** The speed (m/min) of the multifilament-winding roll unit refers to the length by which the plurality of raw filaments are wound on the multifilament-winding roll unit per unit time.

**[0184]** The speed (m/min) of the receiving roll unit used in the step (C) refers to the length by which the plurality of raw filament are received on the receiving roll unit used in the step (C) ("second receiving roll unit 113" in the case of the first embodiment) per unit time.

<sup>35</sup> **[0185]** The speed (m/min) of the stretching roll unit refers to the length by which the plurality of raw filaments are transferred by the stretching roll unit per unit time.

**[0186]** In the first embodiment, only one stretching roll unit is used. A plurality of stretching roll units may be used. When a plurality of stretching roll units are used, the highest of the speeds of the stretching roll units is adopted as the "speed of stretching roll unit".

**[0187]** In the step (B) of FIG. 1, the raw filaments 100A received on the first receiving roll unit 107 are wound on the raw filament-winding roll unit 112. Alternatively, in the first embodiment, the raw filaments 100A received on the first receiving roll unit 107 may be placed into a container without being wound on the raw filament-winding roll unit 112.

**[0188]** In the case where the raw filaments 100A received on the first receiving roll unit 107 are transferred to a container with the aid of a transfer roll unit without being wound on the raw filament-winding roll unit 112, the stretching ratio in the step (B) can be determined by the following equation.

[0189] Stretching ratio = speed (m/min) of transfer roll unit/speed (m/min) of first receiving roll unit

**[0190]** The speed (m/min) of the transfer roll unit refers to the length by which the raw filaments are transferred by the transfer roll unit per unit time.

**[0191]** When a plurality of transfer roll units are used, the highest of the speeds of the transfer roll units is used in the equation for determining the stretching ratio.

[0192] When neither the raw filament-winding roll unit 112 nor any transfer roll unit is used, the stretching ratio is 1.0.

<Second Embodiment: Spin Draw Process>

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55 [0193] Hereinafter, a second embodiment will be described with reference to FIG. 3.

**[0194]** Descriptions already given in the first embodiment are omitted. Features that are not described in the second embodiment should be understood to be the same as those described in the first embodiment.

[0195] A multifilament production method according to the second embodiment is a method in which a spin draw

process is used to produce the multifilament.

**[0196]** The spin draw process is a process in which the step of discharging a melt from the plurality of discharge holes to obtain a plurality of raw filaments in a molten state and the step of stretching the plurality of raw filaments by a stretching roll unit are carried out in one stage. The spin draw process is also called "SDY process" or "direct spin draw process".

[0197] In the step (C) of the second embodiment, as shown in FIG. 3, the plurality of raw filaments cooled in the step (B) are received on a receiving roll unit 207.

**[0198]** Next, the plurality of raw filaments received on the receiving roll unit 207 are stretched by three stretching roll units (a first stretching roll unit 208, a second stretching roll unit 209, and a third stretching roll unit 210).

[0199] In the step (C), the multifilament obtained by the stretching is wound on a multifilament-winding roll unit 212.

[0200] In the step (C), the raw filaments 100A stretched by the stretching roll units may be transferred by a take-off roll unit 211.

[0201] In the step (C), the plurality of raw filaments cooled in the first and second cooling boxes 105 and 106 are received on the receiving roll unit 207.

[0202] The receiving roll unit 207 includes two rolls in FIG. 1, but may include one roll or three or more rolls.

[0203] In the second embodiment, the stretching roll units may serve as heat treatment roll units.

[0204] Each of the stretching roll units 208, 209, and 210 (each of the heat treatment roll units 208, 209, and 210) includes two rolls in FIG. 1, but may include one roll or three or more rolls.

[0205] In terms of accelerating the crystallization of the polymer component contained in the plurality of raw filaments 100A, the temperature of the heat treatment roll units is preferably from 25 to 100°C and more preferably from 40 to 90°C.

**[0206]** When the temperature of the environment where the step (C) is performed is 25°C or above, the crystallization of the polymer component contained in the plurality of raw filaments 100A can be accelerated without using any heat treatment roll unit.

**[0207]** In the multifilament production method according to the present embodiment, the spinning draft ratio (NDR) is preferably 150 or more and more preferably 200 or more. The NDR is usually 1000 or less.

**[0208]** The NDR can be determined by the following equation.

NDR = speed (m/min) of receiving roll unit (first receiving roll unit) that first

receives raw filaments from spinning nozzle/spinning nozzle flow speed (m/min)

**[0209]** When the NDR is 150 or more, the raw filaments can be stretched between the spinning nozzle and the first receiving roll unit, and consequently the strength of the multifilament can be increased.

[0210] In the first embodiment (post-stretching process), the first receiving roll unit is the first receiving roll unit 107.

[0211] In the second embodiment (spin draw process), the first receiving roll unit is the receiving roll unit 207.

<Staple Production Method>

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**[0212]** In a staple production method according to the present embodiment, the multifilament production method according to the present embodiment is used to obtain the multifilament, and the multifilament is cut to obtain a staple.

[0213] The staple has a crimped structure.

**[0214]** The staple has an average length of 200 mm or less, and the average length is preferably 160 mm or less and in particular from 1.0 to 100 mm.

**[0215]** In the staple production method according to the present embodiment, the multifilament may be subjected to crimping, and the multifilament subjected to the crimping may be cut to obtain a staple having a crimped structure.

**[0216]** The multifilament, the multifilament production method, the staple, and the staple production method according to the present embodiment have the features as described above and thus offer the following advantages.

**[0217]** That is, the multifilament according to the present embodiment includes a plurality of individual filaments. The individual filaments contain a poly(3-hydroxyalkanoate) resin and polycaprolactone. The individual filaments have a fineness of 1.0 to 5.0 dtex. The individual filaments have a tensile strength of 2.5 cN/dtex or more.

[0218] Thanks to these features, the multifilament according to the present embodiment exhibits high strength.

**[0219]** In terms of applicability for various purposes, a demand for a multifilament having high strength, including thin individual filaments, and having high biodegradability can arise in the feature.

**[0220]** The polycaprolactone, like the poly(3-hydroxyalkanoate) resin, has biodegradability.

[0221] The individual filaments having a fineness of 5.0 dtex or less are thin.

**[0222]** Thus, according to the present embodiment, a multifilament having high strength, including thin individual filaments, and having high biodegradability can be provided.

[0223] The multifilament production method according to the present embodiment is a method for producing a multi-

filament including a plurality of individual filaments by using a spinning nozzle having a plurality of discharge holes.

**[0224]** The multifilament production method according to the present embodiment includes the steps of: (A) discharging a melt from the plurality of discharge holes to obtain a plurality of raw filaments in a molten state; (B) blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments in the molten state to cool the plurality of raw filaments; and (C) stretching the plurality of cooled raw filaments by a stretching roll unit to obtain the multifilament.

**[0225]** The melt contains a poly(3-hydroxyalkanoate) resin and polycaprolactone. The individual filaments have a fineness of 1.0 to 5.0 dtex.

[0226] In the step (B), cooling the raw filaments by the gas having a temperature of 3 to 15°C can shorten the time during which the temperature of the raw filaments is in a temperature range where the polycaprolactone and poly(3-hydroxyalkanoate) resin constituting the raw filaments crystallize, and thus can retard the crystallization of the polycaprolactone and poly(3-hydroxyalkanoate) resin. This can reduce the degree of hardening of the raw filaments. Thus, the raw filaments are easy to stretch satisfactorily in the step (C). As such, the strength of the multifilament can be easily increased.

**[0227]** In the multifilament production method according to the present embodiment, in the step (C), the plurality of stretched raw filaments are heated by a heat treatment roll unit to obtain the multifilament.

**[0228]** Thanks to this feature, the multifilament production method according to the present embodiment can increase the crystallinity of the multifilament and therefore the strength of the multifilament.

**[0229]** Further, in the multifilament production method according to the present embodiment, the plurality of raw filaments are cooled to 50°C or below in the step (B) and, in the step (C), the plurality of raw filaments cooled to 50°C or below in the step (B) are heated, and the plurality of heated raw filaments are stretched by the stretching roll unit.

**[0230]** To increase the degree of orientation of the polymer component, it is generally desired to stretch the raw filaments in a temperature range suitable for increasing the degree of orientation of the polymer component. This is because, if the raw filaments are stretched at a temperature above the temperature range, the polymer component is brought into a molten state and thus the stretching does not increase the degree of orientation of the polymer component very much. Another reason is that if the raw filaments are stretched at a temperature below the temperature range, the polymer component becomes so hard as to make stretching of the raw filaments difficult and that in this case, forcibly pulling the raw filaments to stretch the raw filaments could cause breakage of the raw filaments, leading to a failure to produce the multifilament.

**[0231]** In the present embodiment, the plurality of raw filaments cooled to 50°C or below in the step (B) are heated, and the plurality of heated raw filaments are stretched by the stretching roll unit. Thus, the temperature of the plurality of raw filaments to be stretched is easier to control within a temperature range suitable for increasing the degree of orientation of the polymer component than in the case of a spin draw process in which a plurality of raw filaments are stretched while being cooled by ambient air. As such, the degree of orientation of the polymer component of the plurality of raw filaments can be easily increased.

[0232] Thus, in the present embodiment, the strength of the multifilament can be easily increased.

**[0233]** The multifilament, the multifilament production method, the staple, and the staple production method according to the present invention are not limited to the embodiment described above. The multifilament, the multifilament production method, the staple, and the staple production method according to the present invention are not limited by the advantageous effects described above. The multifilament, the multifilament production method, the staple, and the staple production method according to the present invention may be modified in various ways without departing from the gist of the present invention.

Examples

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45 [0234] Hereinafter, the present invention will be described in more detail using Examples and Comparative Examples. The present invention is not limited by these Examples in any respect.

<Example 1>

50 [0235] A multifilament was made by the method of the first embodiment (post-stretching process).

(Step (A))

**[0236]** First, as shown in FIG. 1, a melt prepared by the kneading extruder 102 is discharged from the discharge holes of the spinning nozzle 104 to obtain a plurality of (80) raw filaments 100A.

**[0237]** The melt is a polymer composition containing the following components in the proportions shown in Table 1 below: (3-hydroxybutyrate-co-3-hydroxyhexanoate) copolymer resin (P3HB3HH) as a poly(3-hydroxyalkanoate) resin (3-hydroxyhexanoate content = 6 mol%, Mw =  $55 \times 10^4$ ); polycaprolactone (PCL); erucamide as a lubricant having an

amide bond; behenamide as a lubricant having an amide bond; and pentaerythritol as a nucleating agent.

**[0238]** In Table 1 below, "Amount of lubricant having amide bond" refers to the amount (parts by mass) of the lubricant having an amide bond per 100 parts by mass of the P3HB3HH. "Amount of nucleating agent" refers to the amount (parts by mass) of the nucleating agent per 100 parts by mass of the P3HB3HH. Additionally, "parts by mass" is simply written as "parts".

(Step (B))

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[0239] In the cooling boxes 105 and 106, air having a temperature of 7°C (quench air) was blown onto the plurality of raw filaments 100A.

**[0240]** Subsequently, the plurality of raw filaments 100A cooled in the cooling boxes 105 and 106 were received on the first receiving roll unit 107, passed through the transfer roll units 108, 109, 110, and 111 in succession, and then wound on the raw filament-winding roll unit 112.

[0241] The fineness of the raw filaments wound on the raw filament-winding roll unit 112 was 7.6 dtex.

5 **[0242]** The NDR was 353 and the stretching ratio was 1.1.

(Step (C))

**[0243]** As shown in FIG. 2, the plurality of raw filaments (ordinary temperature (25°C)) wound on the raw filament-winding roll unit 112 were received on the second receiving roll unit 113 (ordinary temperature (25°C)) and stretched by the stretching roll unit (heat treatment roll unit) 114 (60°C). The plurality of stretched raw filaments were passed through the take-off roll unit (heat treatment roll unit) 115 (60°C), and then the resulting multifilament was wound on the multifilament-winding roll unit 116.

[0244] The stretching ratio was 2.2 and the relaxation rate was 10%.

(Example 2)

[0245] A multifilament was made by the method of the second embodiment (SDY process).

**[0246]** A plurality of raw filaments 100A cooled in the cooling boxes 105 and 106 were obtained in the same manner as in Example 1.

**[0247]** Subsequently, as shown in FIG. 3, the plurality of raw filaments 100A cooled in the cooling boxes 105 and 106 were received on the receiving roll unit 207 and stretched by the stretching roll units (heat treatment roll units) 208, 209, and 210 (50°C). The plurality of stretched raw filaments were passed through the take-off roll unit 211, and then the resulting multifilament was wound on the multifilament-winding roll unit 212.

[0248] The NDR was 324, the stretching ratio was 2.0, and the relaxation rate was 5%.

(Examples 3, 5, and 7 and Comparative Example 1)

[0249] Multifilaments were obtained in the same manner as the multifilament of Example 1, except that the conditions were changed as shown in Table 1.

(Examples 4, 6, and 8 and Comparative Example 2)

**[0250]** Multifilaments were obtained in the same manner as the multifilament of Example 2, except that the conditions were changed as shown in Table 1.

(Fineness and Tensile Strength of Individual Filaments)

**[0251]** For each of the multifilaments of Examples and Comparative Examples, the fineness and tensile strength of the individual filaments of the multifilament were measured by the methods previously described.

[0252] The values of the fineness and tensile strength of the individual filaments are shown in Table 1 below.

(Matrix-Domain Structure of Individual Filaments)

<sup>55</sup> **[0253]** Whether the individual filaments of each of the multifilaments of Examples and Comparative Examples had a matrix-domain structure was examined by means of a transmission electron microscope (TEM).

**[0254]** The individual filaments of each of the multifilaments of Examples and Comparative Examples were stained with ruthenium tetroxide, and the stained individual filaments were observed with the transmission electron microscope.

Polymers contained in the matrix and domain were identified based on the difference in staining state.

**[0255]** The presence or absence of a matrix-domain structure in the individual filaments, the polymer contained in the matrix, and the polymer contained in the domain are shown in Table 1 below.

5 (Degree of Elongation and Young's Modulus of Individual Filaments)

**[0256]** For each of the multifilaments of Examples and Comparative Examples, the degree of elongation and Young's modulus of the individual filaments of the multifilament were measured at an initial length of 20 mm and a speed of 20 mm/min based on JIS L 1015:2010 "Test methods for man-made staple fibers".

10 [0257] The values of the degree of elongation and Young's modulus of the individual filaments are shown in Table 1 below.

(Fusion Percentage)

15 [0258] For each of the multifilaments of Examples and Comparative Examples, the multifilament was cut along a plane perpendicular to the longitudinal direction of the multifilament, and thus all of the individual filaments of the multifilament were cut.

**[0259]** Subsequently, the cut surface of the multifilament was observed with a scanning electron microscope (SEM), and the total number of individual filaments of the multifilament in the cut surface and the number of individual filaments fused with other individual filaments in the cut surface ("the total number of individual filaments of the multifilament" minus "the number of individual filaments not fused with any other individual filaments") were counted.

[0260] The fusion percentage was determined by the following equation.

Fusion percentage (%) = (number of individual filaments fused with other individual filaments in cut surface/total number of individual filaments of multifilament in cut surface)  $\times$  100

30 **[0261]** The values of the fusion percentage are shown in Table 1 below.

(Heat Resistance Test)

**[0262]** For each of the multifilaments of Examples and Comparative Examples, the individual filaments of the multifilament were subjected to thermomechanical analysis (TMA), and the temperatures at which the shrinkage percentage of the individual filaments reached 5%, 10%, and 20% were measured.

Load: 3 q

Measurement atmosphere: Air

Temperature range: From room temperature (25°C) to 180°C

Temperature rise rate: 5°C/min

[0263] The results of the heat resistance test (the temperatures at which the shrinkage percentage reached 5%, 10%, and 20%) are shown in Table 1 below.

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# [Table 1]

55		B Jymer compasition (sucto								35	e dirions	)f steps				Maltiflament								
									Step (B)			Step (C)												
50		Blend ratio between polymer components	Amount of Inbricant having amide bond	Amonnt of nucleating agent	Weight-average malecular weight of P3HB3HB (Mw)	Weight-average molecular weight of PCL (Mw)	Production method	NDR (-)	Temperature of quench sir (°C)	Stretching ratio (-)	Bueness of raw filaments (dtex)	Streiching ratio (-)	Temperature of heat-treatment roll unit (°C)	Relaxation vate (%)	Finences of indivisual filaments (dtex)	Tensile strength of individual filaments (cNdtex)	Presence or absence of matrix-domain structure in individual filaments	Polymer contained in matrix	Potymer contained in domain	Degree of clongation of individual filaments (%)	Young's modulus of individual filaments (GPa)	Fusion percentage (%)	Beat resistance test (Femperatures at which shrinkage percentage reached 5%, 10%, and 20% (C))	Company of the compan
40	Bx.1	P3HB3HHPCL 60/40 (mass vatio) 55/45 (volume ratio)	Erncamide 0.5 parts Behenamide 0.5 parts	Pentavrythritol 1.0 parts	100.002	70,800	Post-stretching	353	+	1.1	7.6	2.2	09	10	3.6	3.02	Present	РЗИВЗИН	PCI.	57.8	6.1	25.4	58.3 (5%) 61.4 (10%) 85.5 (20%)	101 101 101
40	Ex. 2	P3HB3HH/PCL 60/40 (mass ratio) 55/45 (votume ratio)	Frucamide 0.5 parts Bekenamide 0.5 parts	Pentaerythritol 1.0 paxts	900,008	70,000	SDY	324	7	1	-	2.0	95	v	3.5	2.50	Present	РЗИВЗИН	PCL	73.4	1.7	21.1	57.9 (5%) 63.5 (10%) 84.4 (20%)	(a) (a) (b)
35	Fx. 3	F3HB3HH/PCL 70/30 (mass ratio) 68/35 (volume ratio)	Eracamide 0.5 parts Behenamide 0.5 parts	Pentaerythritol 1.0 parts	500,000	70,000	Post-stretching	236	ė	1.1	9.5	2.3	95	91	4.3	2.60	Present	рэнвэни	PCL	8.99	1.8	23.4	54,4 (5%) 57,5 (10%) 86,3 (20%)	(a : a = )a.a
30	Ex. 4	P3HB3HH/PCI, 70/30 (mass ratio) 65/35 (volume ratio)	Encamide 0.5 parts Behenamide 0.5 parts	Pentaerytbritol 1.0 parts	500.000	70,006	SDY	236	t-	4	-	2.2	80	\$	4.3	2.50	Present	езивзии	PCL	88.2	1.6	22.8	52.3 (5%) 55.5 (10%) 79.7 (26%)	64. Ami 116.1
25	Ex. 5	P3HB3HH/PCJ. 60/40 (mass ratio) 55/45 (volume ratio)	Erucamide 0.5 parts Bebenamide 0.5 parts	Pentaerythritol 1.0 parts	350,000	76.000	Post-stretching	206	٦	1.3	10.9	2.7	69	16	3.6	2.83	Present	РЗНВЗНН	PCI.	41.3	2.4	20.8	51.2 (5%) 56.8 (10%) 78.8 (20%)	introduction .
20	Fx. 6	P3HB3HH/PCL 60/40 (mass ratio) 55/45 (votame ratio)	Erucamide 0.5 parts Behenamide 0.5 parts	Pentaerythritol 1.8 parts	350,000	70,640	SDY	266	7	-	1	2.4	50	s	4.4	2.60	Present	РЗНВЗИН	BCL	84.6	61	23.9	51.2 (5%) 57.9 (10%) 73.7 (20%)	(***)
15	Ex. 7	P3HB3HH/PCL 50/50 (mass ratio) 45/5\$ (volume ratio)	Erucamide 0.5 parts Behenamide 0.5 parts	Pentaerythritol 1.0 parts	900,005	70,990	Post-stretching	236	7	1.1	1.6	2.8	60	10	3.2	3,99	Prescut	PCI,	ннеянеа	38.0	3.9	26.4	48.5 (5%) 53.3 (10%) 68.2 (20%)	
45	Fx. 8	P3HB3HH/PCL 50/50 (mass ratio) 45/55 (volume ratio)	Fracamide 0.5 parts Behenamide 0.5 parts	Peutacrythritol 1.0 parts	500,006	70,000	sox	236	7	ı	ı	2.0	30	s	2.9	2,62	Present	PC1.	езпвзнн	71.6	1,4	12.4	47.5 (5%) 54.4 (10%) 67.8 (20%)	2
10	Сощр. Ух. 1	рзнвзни	Ern camide 0.5 parts Behenamide 0.5 parts	Pentacrythritol 1.0 parts	350,000		Post-stre tching	891	10	1.05	w	1.96	36	10	2.2	2.14	Absent		,	52.5	2.3	30.8	70.3 (5%) 86.5 (10%) 100.1 (20%)	Car can venny
5	Comp. Ex. 2	РЗЯВЗНН	Fracamide 0.5 parts Behenamide 0.5 parts	Pentacrythrifol 1.6 parts	350,000	1	λαs	236	16	]	I	1.5	80	16	2.2	2,00	Absent			80.8	2,4	31.2	71.3 (\$%) 82.5 (10%) 106.3 (20%)	in the second
			_																					

[0264] As seen from Table 1, the tensile strength of the individual filaments was higher in Examples 1 to 8 falling within the scope of the present invention than in Comparative Examples 1 and 2 in which polycaprolactone was not used.

[0265] This demonstrates that the present invention allows for obtaining a multifilament having high strength.

**[0266]** As seen from Table 1, the temperatures at which the given shrinkage percentages were reached were higher in Examples 1 to 6 in which the matrix contained a poly(3-hydroxyalkanoate) resin than in Examples 7 and 8 in which the matrix contained polycaprolactone.

[0267] This demonstrates that a multifilament having a matrix containing a poly(3-hydroxyalkanoate) resin has high heat resistance.

(Comparative Example 3)

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**[0268]** An attempt was made to make a multifilament in the same manner as in Example 1, except that the temperature of quench air was 25°C. The raw filaments broke between the spinning nozzle 104 and the first receiving roll 107, and any multifilament was not able to be made.

(Comparative Example 4)

**[0269]** An attempt was made to make a multifilament in the same manner as in Example 3, except that the temperature of quench air was 25°C. The raw filaments broke between the spinning nozzle 104 and the first receiving roll 107, and any multifilament was not able to be made.

(Comparative Example 5)

**[0270]** An attempt was made to make a multifilament in the same manner as in Example 7, except that the temperature of quench air was 25°C. The raw filaments broke between the spinning nozzle 104 and the first receiving roll 107, and any multifilament was not able to be made.

**[0271]** As described above, in Comparative Examples 3 to 5 in which the temperature of quench air was 25°C, the raw filaments broke between the spinning nozzle 104 and the first receiving roll 107, and any multifilament was not able to be produced.

[0272] In Comparative Examples 3 to 5, multifilaments could be produced if the NDR is reduced. However, reducing the NDR allows the polymer component to crystalize to a certain extent while stretching of the individual filaments is restricted.

**[0273]** In Examples 1 to 8 falling within the scope of the present invention, the step (B), where a gas having a temperature of 3 to 15°C is blown onto a plurality of raw filaments in a molten state, allows the raw filaments to be stretched to a certain extent before crystallization of the polymer component. This is believed to have resulted in multi filaments having high strength.

[0274] Thus, it has been demonstrated that the present invention allows for obtaining a multifilament having high strength.

#### 40 Reference Signs List

## [0275]

100A: raw filament, 101: material feeder, 102: kneading extruder, 103: gear pump, 104: spinning nozzle, 105: first cooling box, 106: second cooling box, 107: first receiving roll unit, 108: first transfer roll unit, 109: second transfer roll unit, 110: third transfer roll unit, 111: fourth transfer roll unit, 112: raw filament-winding roll unit, 113: second receiving roll unit, 114: stretching roll unit (heat treatment roll unit), 115: take-off roll unit (heat treatment roll unit), 116: multifilament-winding roll unit

207: receiving roll unit, 208: first stretching roll unit (heat treatment roll unit), 209: second stretching roll unit (heat treatment roll unit), 210: third stretching roll unit (heat treatment roll unit), 211: take-off roll unit, 212: multifilament-winding roll unit

#### Claims

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1. A multifilament comprising a plurality of individual filaments, wherein

the individual filaments contain a poly(3-hydroxyalkanoate) resin and polycaprolactone,

the individual filaments have a fineness of 1.0 to 5.0 dtex, and the individual filaments have a tensile strength of 2.5 cN/dtex or more.

- 2. The multifilament according to claim 1, wherein a mass ratio of the poly(3-hydroxyalkanoate) resin to the polycaprolactone is from 30/70 to 80/20.
  - **3.** The multifilament according to claim 1 or 2, wherein the poly(3-hydroxyalkanoate) resin contains a structural unit represented by the following formula (1):

[-CHR-CH<sub>2</sub>-CO-O-] (1), wherein R is an alkyl group represented by  $C_pH_{2p+1}$  and p is an integer from 1 to 15.

4. The multifilament according to any one of claims 1 to 3, wherein

the individual filaments have a matrix-domain structure including a matrix and a domain, the matrix contains the poly(3-hydroxyalkanoate) resin, and the domain contains the polycaprolactone.

5. A staple comprising a cut piece of the multifilament according to any one of claims 1 to 4, wherein

the staple has a crimped structure, and the staple has an average length of 200 mm or less.

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**6.** A multifilament production method for producing a multifilament including a plurality of individual filaments by melt spinning using a spinning nozzle having a plurality of discharge holes, the multifilament production method comprising the steps of:

(A) discharging a melt from the plurality of discharge holes to obtain a plurality of raw filaments in a molten state;

- (B) blowing a gas having a temperature of 3 to 15°C onto the plurality of raw filaments in the molten state to cool the plurality of raw filaments; and
- (C) stretching the plurality of cooled raw filaments by a stretching roll unit to obtain the multifilament, wherein

the melt contains a poly(3-hydroxyalkanoate) resin and polycaprolactone, and the individual filaments have a fineness of 1.0 to 5.0 dtex.

- 7. The multifilament production method according to claim 6, wherein the individual filaments have a tensile strength of 2.5 cN/dtex or more.
  - **8.** The multifilament production method according to claim 6 or 7, wherein in the step (C), the plurality of stretched raw filaments are heated by a heat treatment roll unit to obtain the multifilament.
- **9.** The multifilament production method according to any one of claims 6 to 8, wherein a stretching ratio in the step (C) is 2.0 or more.
  - 10. The multifilament production method according to any one of claims 6 to 9, wherein
- in the step (B), the plurality of raw filaments are cooled to 50°C or below, and in the step (C), the plurality of raw filaments cooled to 50°C or below in the step (B) are heated, and the plurality of heated raw filaments are stretched by the stretching roll unit.
  - **11.** The multifilament production method according to claim 10, wherein the raw filaments used in the step (C) have a fineness of 5.0 to 15.0 dtex.
  - 12. The multifilament production method according to claim 10 or 11, wherein in the step (C), the plurality of raw filaments cooled in the step (B) are received on and heated by a receiving roll unit having a temperature that is 25°C or above but below 50°C, the plurality of raw filaments heated by the receiving roll unit are stretched by the stretching roll unit, and the plurality of raw filaments stretched by the stretching roll unit are heated by a heat treatment roll unit having a temperature of 40 to 100°C.
  - 13. The multifilament production method according to any one of claims 6 to 12, wherein

in the step (B), the plurality of cooled raw filaments are wound on a raw filament-winding roll unit, and in the step (C), the plurality of raw filaments wound on the raw filament-winding roll unit are stretched by the stretching roll unit.

- **14.** The multifilament production method according to any one of claims 6 to 9, wherein a spin draw process is used to produce the multifilament.
  - **15.** The multifilament production method according to claim 14, wherein in the step (C), the plurality of raw filaments stretched by the stretching roll unit are heated by a heat treatment roll unit having a temperature of 25 to 100°C.
  - **16.** A staple production method comprising:

using the multifilament production method according to any one of claims 6 to 15 to obtain the multifilament; and cutting the multifilament to obtain a staple, wherein

the staple has a crimped structure, and

the staple has an average length of 200 mm or less.

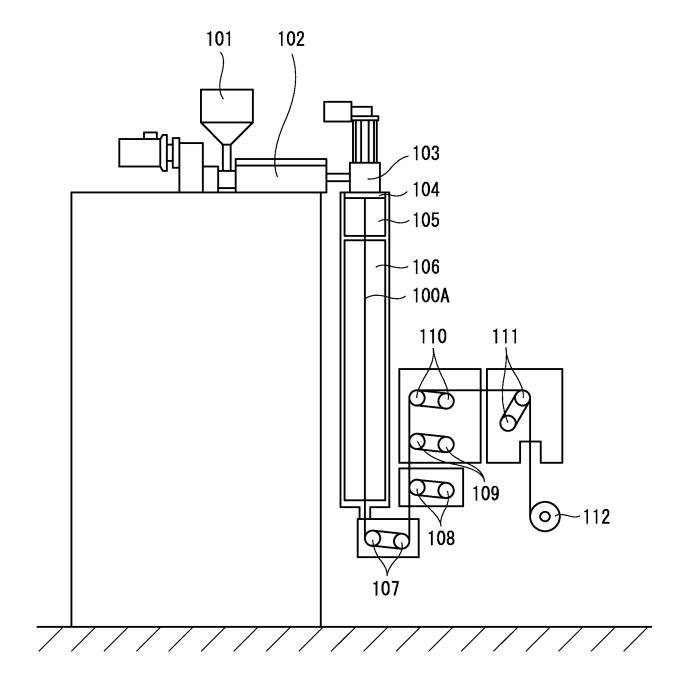
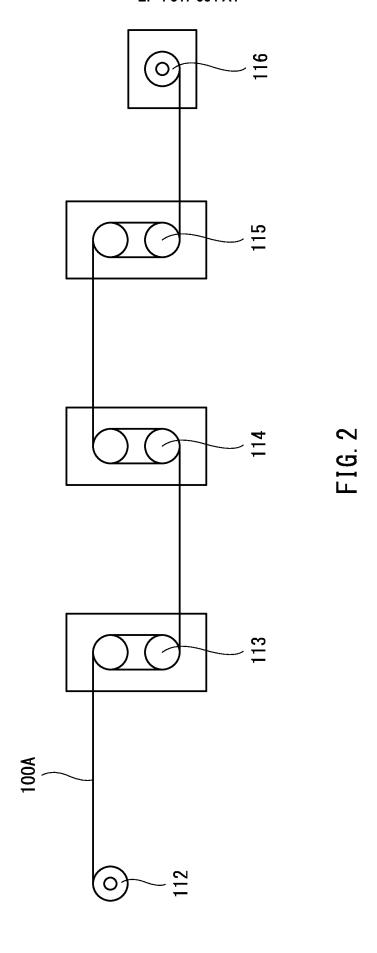


FIG. 1



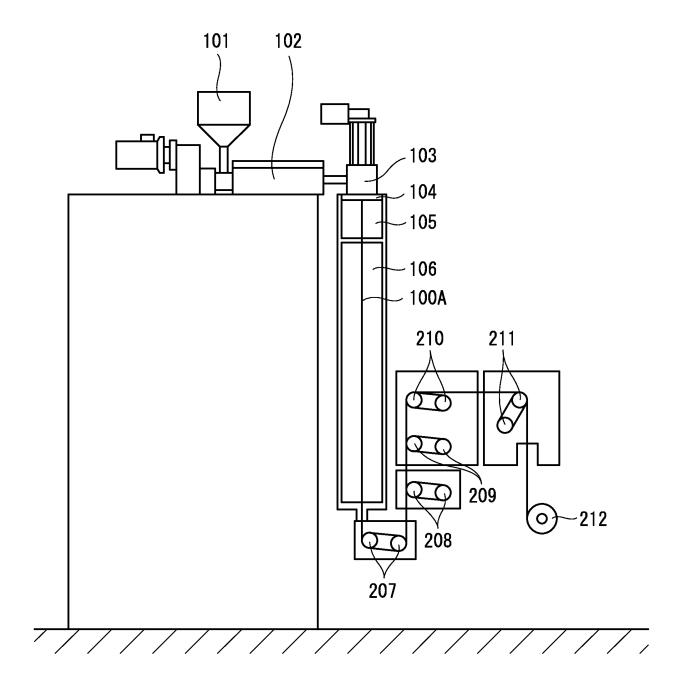


FIG. 3

International application No.

INTERNATIONAL SEARCH REPORT

#### PCT/JP2022/010854 5 CLASSIFICATION OF SUBJECT MATTER *D02J 1/22*(2006.01)i; *D02J 13/00*(2006.01)i; *D01F 6/92*(2006.01)i FI: D01F6/92 307A; D02J1/22 J; D02J13/00 S According to International Patent Classification (IPC) or to both national classification and IPC 10 В. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) D02J1/00-1/22; D02J13/00; D01F1/00-13/04 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Published examined utility model applications of Japan 1922-1996 15 Published unexamined utility model applications of Japan 1971-2022 Registered utility model specifications of Japan 1996-2022 Published registered utility model applications of Japan 1994-2022 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) 20 C. DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Category\* JP 6-264305 A (UNITIKA LTD) 20 September 1994 (1994-09-20) X 1-5 claims 1-3, paragraph [0001], table 1, examples 2-4 25 Α claims 1-3, paragraph [0001], table 1, examples 2-4 6-16 X JP 5-093316 A (UNITIKA LTD) 16 April 1993 (1993-04-16) 1-3, 5-16 claims 1-2, paragraphs [0001], [0016]-[0020], table 1, examples 1-3 Α claims 1-2, paragraphs [0001], [0016]-[0020], table 1, examples 1-3 4 Α JP 5-093317 A (UNITIKA LTD) 16 April 1993 (1993-04-16) 1-16 30 claims 1-2, example 1 US 2010/0057123 A1 (ANGIOTECH PHARMACEUTICALS, INC.) 04 March 2010 1-16 Α (2010-03-04)claims 1-20 35 Further documents are listed in the continuation of Box C. See patent family annex. later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance 40 document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step earlier application or patent but published on or after the international filing date when the document is taken alone document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art document referring to an oral disclosure, use, exhibition or other 45 document member of the same patent family document published prior to the international filing date but later than the priority date claimed $\,$ Date of the actual completion of the international search Date of mailing of the international search report 31 May 2022 17 May 2022 50 Name and mailing address of the ISA/JP Authorized officer Japan Patent Office (ISA/JP) 3-4-3 Kasumigaseki, Chiyoda-ku, Tokyo 100-8915 Japan Telephone No.

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International application No.

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#### REFERENCES CITED IN THE DESCRIPTION

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