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(71) Applicants:
• **ES FiberVisions Co., Ltd.**
Osaka-shi, Osaka 530-6108 (JP)

- **ES Fibervisions Hong Kong Limited**
Kowloon, Hong Kong (CN)
- **ES FiberVisions LP**
Athens, Georgia 30601 (US)
- **ES Idorama Ventures Hygiene Denmark ApS**
6800 Varde (DK)

(72) Inventor: **WADA, Tomoyuki**
Osaka-shi, Osaka 530-6108 (JP)

(74) Representative: **Becker, Eberhard**
Becker Kurig & Partner
Patentanwälte mbB
Bavariastraße 7
80336 München (DE)

(54) **HEAT-FUSIBLE COMPOSITE FIBER AND NON-WOVEN FABRIC USING SAME**

(57) The purpose of the present invention is to provide a heat-fusible composite fiber that reduces the consumption of fossil resources and imparts both bulkiness and flexibility to non-woven fabrics, and a non-woven fabric using the heat-fusible composite fiber. Provided is a heat-fusible composite fiber in which a first component

is configured from a polyester resin and a second component is configured from a polyethylene resin having a lower melting point than the first component, the mixing ratio (weight ratio) of a biomass-derived polyethylene resin and a fossil-resource-derived polyethylene resin in the polyethylene resin being 20:80 to 90: 10.

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Description

[Technical Field]

5 **[0001]** The present invention relates to a heat-fusible composite fiber containing a biomass-derived component, and a non-woven fabric obtained using the same.

[Background Art]

10 **[0002]** In the related art, heat-fusible composite fibers, which can be formed by heat fusion using the hot air current or heat energy from heating rolls, made it easy to obtain non-woven fabrics having excellent bulkiness and flexibility, and thus have been widely used for sanitary materials such as diapers, napkins, and pads; daily necessities; or industrial materials such as filters. The importance of bulkiness and flexibility is extremely high particularly in sanitary materials because they come in direct contact with human skin and are required to quickly absorb liquids such as urine and
15 menstrual blood. In order to obtain bulkiness, an approach of using a highly rigid resin or an approach of imparting rigidity by drawing at a high ratio is typical, but in that case, the flexibility of the obtained non-woven fabric decreases. On the other hand, when flexibility is prioritized, the bulkiness of the obtained non-woven fabric becomes low, making liquid absorbency poor.

20 **[0003]** Therefore, methods have been proposed for obtaining fibers and non-woven fabrics capable of achieving both bulkiness and flexibility. Patent Literature 1 discloses a heat-fusible composite fiber in which a first component is a polyester resin and a second component is a polyolefin resin having a lower melting point than that of the first component, and discloses that a bulky and flexible non-woven fabric is obtained using this fiber.

25 **[0004]** Meanwhile, in recent years, along with the increasing demand for establishing a recycling-based society, there is a desire to break away from fossil resources in the material field as well as in energy, and the utilization of biomass-derived materials is attracting attention. Biomass is organic compounds that are photosynthesized from carbon dioxide and water (refer to Patent Literature 2 and Patent Literature 3, for example). When such biomass-derived materials are utilized as starting materials, the amount of fossil resources used can be reduced. For example, when a biomass-derived material such as polylactic acid is used as a raw material, even if it is incinerated after use and decomposed into carbon dioxide and water, their amounts are equal to those of carbon dioxide and water before they are taken up by the plant
30 through photosynthesis, making establishing of a recycling system or carbon neutrality possible.

[0005] Against this background, composite fibers made from a biomass-derived material as a raw material have been proposed also in the field of sanitary materials. Patent Literature 4 discloses PET and PE composite fibers made from biomass-derived substances as raw materials, and discloses that non-woven fabrics that reduce the consumption of fossil resources and have a uniform texture by polymerizing PE with various polymers are obtained.

35 **[0006]** Generally, it is thought that the chemical structure of biomass-derived resins is not different from those of conventional fossil resources, and there is no difference in quality therebetween. However, impurities and the like which could not be removed in the production process remain in raw material monomers for biomass-derived resins, resulting in a decrease in heat resistance and the like, which makes them difficult to use as they are in the same manner as fossil-resource-derived resins. In particular, in the production of non-woven fabrics for sanitary materials, there is means of
40 making the fineness of fibers small to obtain favorable flexibility and texture. However, when conventional biomass-derived resins are applied, it is difficult to obtain heat-fusible composite fibers with a small fineness, and even when heat-fusible composite fibers with a small fineness are obtained, non-woven fabrics obtained by using these fibers have a very low bulk.

45 [Citation List]

[Patent Literature]

[0007]

50 [Patent Literature 1]
Japanese Patent Laid-Open No. 2017-214662
[Patent Literature 2]
Japanese Patent Laid-Open No. 2009-091694
55 [Patent Literature 3]
Japanese Patent Laid-Open No. 2008-150759
[Patent Literature 4]
Japanese Patent Laid-Open No. 2012-140728

[Summary of Invention]

[Technical Problem]

5 **[0008]** As described above, in the related art, non-woven fabrics having both bulkiness and flexibility while reducing the consumption of fossil resources have not been obtained because use of materials derived only from fossil resources was required when attempting to obtain non-woven fabrics that achieve both bulkiness and flexibility at satisfactory levels for suitable use as sanitary materials.

10 **[0009]** The present invention has been made against the background of the above-mentioned related art, and an objective thereof is to provide a heat-fusible composite fiber that reduces the consumption of fossil resources and imparts both bulkiness and flexibility to non-woven fabrics, and a non-woven fabric using the heat-fusible composite fiber.

[Solution to Problem]

15 **[0010]** In order to achieve the above-mentioned objective, the inventors of the present invention have made extensive research. As a result, it has been found that the above-mentioned objective can be achieved by mixing a biomass-derived polyethylene resin and a fossil-resource-derived polyethylene resin in an appropriate ratio as a polyethylene resin in a heat-fusible composite fiber in which a first component is configured from a polyester resin and a second component is configured from a polyethylene resin having a lower melting point than that of the first component, thereby
20 completing the present invention.

[0011] That is, the present invention is configured as follows.

[1] A heat-fusible composite fiber containing: a first component configured from a polyester resin; and a second component configured from a polyethylene resin having a lower melting point than that of the first component, in
25 which a mixing ratio (weight ratio) of a biomass-derived polyethylene resin and a fossil-resource-derived polyethylene resin in the polyethylene resin is 20:80 to 90:10.

[2] The heat-fusible composite fiber according to [1], in which a biomass-derived carbon content in the polyethylene resin is 20% to 90%.

30 [3] The heat-fusible composite fiber according to [1] or [2], in which a biomass-derived carbon content in the heat-fusible composite fiber is 10% or more.

[4] The heat-fusible composite fiber according to any one of [1] to [3], in which a biomass-derived carbon content in the polyester resin is 30% or less.

[5] The heat-fusible composite fiber according to any one of [1] to [4], in which a fineness of the heat-fusible composite fiber is 2.2 dtex or less.

35 [6] The heat-fusible composite fiber according to any one of [1] to [5], in which a heat of fusion of the polyester resin in the heat-fusible composite fiber is 24 J/g or more.

[7] The heat-fusible composite fiber according to any one of [1] to [6], in which the heat-fusible composite fiber is a sheath-core type heat-fusible composite fiber containing: the first component as a core component; and the second component as a sheath component.

40 [8] The heat-fusible composite fiber according to any one of [1] to [7], in which the polyester resin is polyethylene terephthalate, and the polyethylene resin is high-density polyethylene.

[9] A non-woven fabric containing the heat-fusible composite fiber according to any one of [1] to [8].

[10] An absorbent article using the heat-fusible composite fiber according to any one of [1] to [8].

45 [Advantageous Effects of Invention]

[0012] According to the present invention, it is possible to provide a heat-fusible composite fiber that reduces the consumption of fossil resources and imparts both bulkiness and flexibility to non-woven fabrics.

50 [Description of Embodiments]

[0013] A heat-fusible composite fiber of the present invention is characterized by containing a first component configured from a polyester resin, and a second component configured from a polyethylene resin having a lower melting point than that of the first component, in which a mixing ratio (weight ratio) of a biomass-derived polyethylene resin and a
55 fossil-resource-derived polyethylene resin in the polyethylene resin is 20:80 to 90:10.

(First component)

[0014] The polyester resin forming the first component in the present invention is not particularly limited, but aromatic polyester resins such as polyethylene terephthalate, polypropylene terephthalate, and polybutylene terephthalate can be preferably used. In addition to the above-mentioned aromatic polyester resins, aliphatic polyester resins can also be used, and examples of preferable aliphatic polyester resins include polylactic acid and polybutylene succinate. These polyester resins may be not only homopolymers but also polyester copolymers (copolyesters). At this time, as copolymer components, it is possible to use dicarboxylic acid components such as adipic acid, sebacic acid, phthalic acid, isophthalic acid, and 2,6-naphthalenedicarboxylic acid; diol components such as diethylene glycol and neopentyl glycol; and optical isomers such as L-lactic acid. Examples of such copolymers include polybutylene terephthalate adipate. Furthermore, two or more of these polyester resins may be mixed and used. Among them, unmodified polymers configured only of polyethylene terephthalate is preferable as the first component in consideration of raw material cost, the bulkiness of non-woven fabrics, the heat stability of obtained fibers, and the like.

[0015] When the polyester resin is an aromatic polyester resin, it can be obtained by condensation polymerization from a diol and a dicarboxylic acid, for example. Examples of dicarboxylic acids used for condensation polymerization of polyester resins include terephthalic acid, isophthalic acid, 2,6-naphthalenedicarboxylic acid, adipic acid, and sebacic acid. Examples of diols used include ethylene glycol, diethylene glycol, 1,3-propanediol, 1,4-butanediol, neopentyl glycol, and 1,4-cyclohexanedimethanol.

[0016] The biomass-derived carbon content in the polyester resin in the present invention is not particularly limited, but is preferably 30% or less, more preferably 2% to 28%, and further preferably 6% to 24%. When the biomass-derived carbon content of the polyester resin is 2% or more, this is preferable because then the consumption of fossil resources can be reduced, and when the biomass-derived carbon content of the polyester resin is 30% or less, this is preferable because then maintaining the original physical properties of the polyester resin becomes easier, which makes it possible to impart bulkiness and flexibility to non-woven fabrics.

[0017] The biomass-derived carbon content is a value obtained by measuring the content of biomass-derived carbon by radioactive carbon (^{14}C) measurement. It is known that, because carbon dioxide in the atmosphere contains a certain percentage of ^{14}C (107 pMC (percent modern carbon)), the ^{14}C content in plants that grow by taking in carbon dioxide in the atmosphere, such as corn, is also about 107 pMC. In addition, ^{14}C returns to nitrogen atoms with a half-life of 5,370 years, and 226,000 years are required to completely decay. Therefore, it is known that, after carbon dioxide in the atmosphere is taken up by plants and the like and fixed, ^{14}C is hardly contained in fossil fuels such as coal, petroleum, and natural gas, which are thought to be over 226,000 years old. Accordingly, the biomass-derived carbon content can be calculated by measuring the percentage of ^{14}C contained in the total carbon atoms in the resin. A method for calculating the biomass-derived carbon content in the resin in the present invention will be described in detail in Examples to be described later.

[0018] The polyester resin is not particularly limited, but incorporating a biomass-derived polyester resin and a fossil-resource-derived polyester resin is preferable from the viewpoint of reducing the consumption of fossil resources and imparting bulkiness and flexibility to non-woven fabrics without impairing the original physical properties of the polyester resin. From this viewpoint, the mixing ratio (weight ratio) of the biomass-derived polyester resin and the fossil-resource-derived polyester resin in the polyester resin is not particularly limited but is preferably 5:95 to 95:5, and more preferably 20:80 to 80:20.

[0019] The biomass-derived polyester resin may contain biomass-derived carbon, and the biomass-derived carbon content is preferably 10% or more, and more preferably 20% or more. Such a biomass-derived polyester resin may be a polymer consisting only of biomass-derived monomers, or may be a copolymer of a biomass-derived monomer and a fossil-resource-derived monomer. For example, when the polyester resin is an aromatic polyester resin, examples thereof include copolymers of biomass-derived diols and biomass-derived dicarboxylic acid, copolymers of biomass-derived diols and fossil-resource-derived dicarboxylic acid, and copolymers of fossil-resource-derived diols and biomass-derived dicarboxylic acid. Among them, copolymers of biomass-derived diols and fossil-resource-derived dicarboxylic acid are preferable from the viewpoint of easy availability.

[0020] The biomass-derived polyester resin is not particularly limited, and a polyester resin obtained by a conventionally known method may be used, or a biomass-derived polyethylene terephthalate commercially available from Far Eastern New Century Corporation, and a biomass-derived polylactic acid commercially available from NatureWorks LLC may also be used.

[0021] Furthermore, the fossil-resource-derived polyester resin means a polyester resin not containing biomass-derived carbon, that is, having a biomass-derived carbon content of 0%. Therefore, the fossil-resource-derived polyester resin is a polyester resin that is polymerized only from fossil-resource-derived monomers.

[0022] The first component is not particularly limited as long as it contains the polyester resin, but preferably contains 80% by mass or more of the polyester resin and more preferably contains 90% by mass or more of the polyester resin. As necessary, an additive such as antioxidants, light stabilizers, ultraviolet absorbers, neutralizing agents, nucleating

agents, epoxy stabilizers, lubricants, antibacterial agents, flame retardants, antistatic agents, pigments, and plasticizers may be added as appropriate within a range not impairing with the effects of the present invention.

(Second component)

[0023] The polyethylene resin in the present invention is not particularly limited. Examples thereof include high-density polyethylene, linear low-density polyethylene, low-density polyethylene, and copolymers of ethylene with other components (α -olefins, for example), and a mixture of these, but the polyethylene resin is preferably configured only of high-density polyethylene from the viewpoint of preventing the phenomenon in which polyethylene resins exposed on a fiber surface are fused together because they are not completely cooled and solidified during spinning.

[0024] In the polyethylene resin forming the second component in the present invention, it is important that the mixing ratio (weight ratio) of a biomass-derived polyethylene resin and a fossil-resource-derived polyethylene resin is 20:80 to 90:10. As in the related art, in a case where only biomass-derived polyethylene resin is used as the polyethylene resin, when the resin is subjected to a heat history of nearly 300°C in the process of melting, this causes the viscosity reduction and the molecular weight reduction of the resin, and sufficient drawability could not be obtained, making it difficult to achieve both small fineness and rigidity of the composite fiber, and therefore, it is thought that flexible and bulky non-woven fabrics cannot be obtained. In the present invention, it has been found that, by setting the mixing ratio of the biomass-derived polyethylene resin to 90% by weight or less, the reduction of the viscosity and the molecular weight of the polyethylene resin is prevented, and extension and elongation in the forming process of the composite fibers becomes an appropriate level, which makes it possible to achieve both small fineness and rigidity of the composite fiber, and furthermore, by setting the mixing ratio of the biomass-derived polyethylene resin to 20% by weight or more, not only the biomass-derived carbon content of the composite fiber can be improved, thereby reducing the consumption of fossil resources, but also the flexibility of non-woven fabrics can be further improved. From this viewpoint, the mixing ratio (weight ratio) of the biomass-derived polyethylene resin and the fossil-resource-derived polyethylene resin is preferably 30:70 to 70:30, and more preferably 40:60 to 50:50.

[0025] The biomass-derived polyethylene resin may contain biomass-derived carbon, and the biomass-derived carbon content is preferably 90% or more, and more preferably 94% or more. Such a biomass-derived polyethylene resin may be a polymer consisting only of biomass-derived monomers, or may be a polymer of a biomass-derived monomer and a fossil-resource-derived monomer. Examples thereof include polymers of biomass-derived ethylene, copolymers of biomass-derived ethylene and biomass-derived α -olefins (propylene, butylene, hexene, octene, and the like), copolymers of biomass-derived ethylene and fossil-resource-derived ethylene, copolymers of biomass-derived ethylene and fossil-resource-derived α -olefins, and copolymers of biomass-derived α -olefins and fossil-resource-derived ethylene. Among them, from the viewpoint of preventing the conglutination between fibers when forming the composite fibers, the biomass-derived polyethylene resin is preferably a polymer of biomass-derived ethylene or a polymer of biomass-derived ethylene and fossil-resource-derived ethylene.

[0026] The biomass-derived polyethylene resin is not particularly limited, and a polyethylene resin obtained by a conventionally known method may be used. For example, the biomass-derived polyethylene resin can be produced by fermenting starch and sugar obtained from corn, sugar cane, sweet potato, and the like with microorganisms to produce bioethanol, dehydrating the bioethanol to produce a biomass-derived ethylene, and polymerizing the biomass-derived ethylene. A biomass-derived polyethylene resin commercially available from Braskem and the like may also be used.

[0027] Furthermore, the fossil-resource-derived polyethylene resin means a polyethylene resin not containing biomass-derived carbon, that is, having a biomass-derived carbon content of 0%. Therefore, the fossil-resource-derived polyethylene resin is a polyethylene resin polymerized only from fossil-resource-derived monomers, and examples thereof include polymers of fossil-resource-derived ethylene, and copolymers of fossil-resource-derived ethylene and fossil-resource-derived α -olefins. Among them, from the viewpoint of preventing the conglutination between fibers when forming the composite fibers, the fossil-resource-derived polyethylene resin is preferably a polymer of fossil-resource-derived ethylene.

[0028] The density of the biomass-derived polyethylene resin is not particularly limited, but examples thereof include 0.91 to 0.96 g/cm³. In addition, the density of the fossil-resource-derived polyethylene resin is not particularly limited, but it is 0.91 to 0.96 g/cm³, for example, and is preferably 0.93 to 0.96 g/cm³ from the viewpoint of expressing appropriate crystallinity and imparting rigidity to the composite fibers.

[0029] The biomass-derived carbon content in the polyethylene resin in the present invention is not particularly limited, but is preferably 20% to 90%, more preferably 30% to 70%, and further preferably 40% to 50%. When the biomass-derived carbon content in the polyethylene resin is 20% or more, this is preferable because this not only reduces the consumption of fossil resources but also imparts flexibility to non-woven fabrics, and when the biomass-derived carbon content in the polyethylene resin is 90% or less, this is preferable because then bulky non-woven fabrics can be obtained.

[0030] The melt mass flow rate (hereinafter abbreviated as MFR) of the polyethylene resin that can be suitably used is not particularly limited, but is preferably 10 to 40 g/10 minutes, more preferably 16 to 20 g/10 minutes, and further

preferably 17 to 19 g/10 minutes. When the MFR of the polyethylene resin is 10 g/10 minutes or more, this is preferable because then stable operability can be obtained, and when the MFR of the polyethylene resin is 40 g/10 minutes or less, this is preferable because then the crystallization of the polyester resin can be promoted, making it possible to obtain bulky non-woven fabrics. The physical properties of the polyethylene resin other than the MFR, such as a Q value (weight-average molecular weight/number-average molecular weight), a Rockwell hardness, and the number of branched methyl chains, are not particularly limited as long as they satisfy the requirements of the present invention.

[0031] The second component is not particularly limited as long as it contains the polyethylene resin, but preferably contains 80% by mass or more of the polyethylene resin and more preferably contains 90% by mass or more of the polyethylene resin. As necessary, additives exemplified for the first component may be contained as appropriate within a range not impairing the effects of the present invention.

(Heat-fusible composite fiber)

[0032] A combination of components forming the heat-fusible composite fiber in the present invention (hereinafter sometimes referred to as "composite fiber") is not particularly limited as long as the first component is configured from the polyester resin, and the second component is configured from the polyethylene resin having a lower melting point than that of the first component, and the combination can be used by selecting from the above-mentioned first component and second component. Specific examples of first component/second component combinations include polyethylene terephthalate/high-density polyethylene, polyethylene terephthalate/linear low-density polyethylene, polyethylene terephthalate/low-density polyethylene, polybutylene terephthalate/high-density polyethylene, and polylactic acid/high-density polyethylene. A preferable combination among these is polyethylene terephthalate/high-density polyethylene.

[0033] The biomass-derived carbon content of the composite fiber in the present invention is not particularly limited, but is preferably 10% or more, more preferably 15% to 60%, and further preferably 25% to 40%. When the biomass-derived carbon content of the composite fiber is 10% or more, this is preferable because then the consumption of fossil resources can be reduced, and when the biomass-derived carbon content of the composite fiber is 60% or less, this is preferable because then maintaining the original physical properties of the resin becomes easier, which makes it possible to impart bulkiness and flexibility to non-woven fabrics.

[0034] The composite fiber of the present invention is not particularly limited, but is preferably a sheath-core heat-fusible composite fiber having the first component as a core component and the second component as a sheath component. Among these, a composite form in which the second component completely covers the surface of the composite fiber is preferable, and a concentric or eccentric sheath-core structure is more preferable. As the cross-sectional shape of the composite fiber, any of a round shape such as circle and ellipse, a square shape such as triangle and quadrangle, an irregular shape such as star and octofoil, and a hollow shape can be used.

[0035] The composition ratio when combining the first component and the second component is not particularly limited, but the first component/the second component is preferably 20/80 to 80/20 (weight ratio), and is more preferably 40/60 to 70/30 (weight ratio). By setting the composition ratio within such a range, the balance between the strength, the bulkiness, and the workability of non-woven fabrics tends to be excellent, which is preferable.

[0036] The fineness of the composite fiber in the present invention is not particularly limited, but is preferably 2.2 dtex or less, more preferably 0.5 to 2.1 dtex, and further preferably 1.6 to 1.8 dtex. When the fineness of the composite fiber is 2.2 dtex or less, it is possible to obtain satisfactory flexibility and texture, especially as non-woven fabrics for sanitary materials.

[0037] The breaking strength of the composite fiber is not particularly limited, but is preferably 1.0 to 4.0 cN/dtex and is more preferably 1.5 to 2.5 cN/dtex for the composite fibers used in absorbent articles, for example. When the breaking strength of the composite fiber is 1.0 cN/dtex or more, non-woven fabrics with sufficient strength can be obtained, and when the breaking strength of the composite fiber is 4.0 cN/dtex or less, the flexibility and the texture of non-woven fabrics can be improved. The breaking elongation of the composite fiber is not particularly limited, but is preferably 30% to 170%, more preferably 50% to 150%, and further preferably 60% to 120%. When the breaking elongation of the composite fiber is 30% or more, this is preferable because then the flexibility and the texture of non-woven fabrics can be improved, and when the breaking elongation of the composite fiber is 170% or less, the rigidity of the composite fiber is increased, which makes it possible to improve the bulkiness of non-woven fabrics.

[0038] The crimp of the composite fiber is not particularly limited, and crimp characteristics such as the presence or absence of crimps, the number of crimps, a crimp rate, a residual crimp rate, and a crimp elastic modulus can be appropriately selected in consideration of web formation methods, the specifications of pieces of web formation equipment, the productivity and the required physical properties of non-woven fabrics, and the like. The shape of the crimp is not particularly limited, and a zigzag-shaped mechanical crimp, a spiral-shaped or ohmic-shaped three-dimensional crimp, or the like can be appropriately selected. Furthermore, the crimp may be visible or hidden in the heat-fusible composite fiber.

[0039] The heat of fusion of the polyester resin in the composite fiber of the present invention is not particularly limited,

but is preferably 24 J/g or more, and more preferably 26 J/g or more. The heat of fusion of the polyester resin in the composite fiber is thought to be a value that reflects the degree of crystallinity of the polyester resin in the composite fiber, and when the heat of fusion is 24 J/g or more, the rigidity of the composite fiber is improved, making it possible to impart bulkiness and flexibility to non-woven fabrics. The upper limit value of the heat of fusion of the polyester resin in the composite fiber is not particularly limited, but is practically 35 J/g or less.

[0040] The fiber length of the heat-fusible composite fiber in the present invention is not particularly limited, but is preferably 3 mm or more, and more preferably 30 to 64 mm. Such a range is preferable because then a web having excellent opening properties and texture is easily obtained in a web formation step such as a carding method, and non-woven fabrics having uniform physical properties can be obtained.

(Method for producing heat-fusible composite fiber)

[0041] A method for producing the heat-fusible composite fiber of the present invention is not particularly limited, and any known method for producing the heat-fusible composite fiber may be employed. Examples of methods for producing the heat-fusible composite fiber with a high productivity and a high yield include a method described later.

(Spinning step)

[0042] A polyester resin, which is a raw material for the composite fiber of the present invention, is allocated to the first component, and a polyethylene resin having a lower melting point than that of the first component is allocated to the second component, thereby forming undrawn fibers in which the first component and the second component are combined by melt-spinning.

[0043] The temperature conditions during melt-spinning are not particularly limited, but the spinning temperature is preferably 250°C or higher, more preferably 280°C or higher, and further preferably 300°C or higher. When the spinning temperature is 250°C or higher, the number of broken yarns during spinning can be reduced, and an undrawn yarn that is likely to retain elongation after drawing can be obtained, thereby easily achieving a small fineness. These effects become more pronounced when the spinning temperature is 280°C or higher, and become further pronounced when the spinning temperature is 300°C or higher, which is preferable. The upper limit of the temperature is not particularly limited as long as it is a temperature at which spinning can be suitably performed.

[0044] The spinning speed is not particularly limited, but is preferably 300 to 1,500 m/minute, and more preferably 400 to 1,000 m/minute. The spinning speed of 300 m/minute or more is preferable from the viewpoint of increasing a single-hole discharge amount when obtaining an undrawn yarn having an arbitrary spinning fineness, thereby obtaining a satisfactory productivity.

(Drawing step)

[0045] The undrawn fibers obtained under the above-mentioned conditions are drawn in a drawing step. The draw temperature is a temperature 30°C to 70°C higher than the glass transition temperature of the polyester resin forming the first component and lower than the melting point of the polyethylene resin forming the second component, and is preferably a temperature 35°C to 60°C higher than the glass transition temperature of the polyester resin and 5°C lower than the melting point of the polyethylene resin.

[0046] The draw temperature means the temperature of the fibers at a draw start position. When the draw temperature is the "glass transition temperature of the polyester resin as the first component + 30°C" or higher, this is preferable because then the effect can be obtained even when drawing at a high strain rate, that is, at a high ratio. The draw temperature is required to be lower than the melting point of the polyethylene resin, which is the second component, to prevent the destabilization of the drawing process due to fusion between fibers. For example, the draw temperature is equal to or higher than 100°C and lower than 130°C when drawing undrawn fibers in which polyethylene terephthalate having a glass transition temperature of 70°C is allocated to the first component, and high-density polyethylene having a melting point of 130°C is allocated to the second component. When the draw temperature is 100°C or higher, the amount of heat with respect to the fibers increases, making the difference in drawability between the polyester resin and the polyethylene resin small. Thus, a risk of causing sheath-core peeling during carding processing in a non-woven fabric formation step is reduced.

[0047] The draw ratio is not particularly limited, but is preferably 2 to 7 times, and more preferably 4 to 6 times. By setting the draw ratio within the above-mentioned range, the small fineness and the rigidity of the composite fiber are well balanced, non-woven fabrics having excellent bulkiness and flexibility can be easily obtained, and furthermore, the composite fiber can be obtained with high productivity.

(Crimp adding step)

[0048] Mechanical crimps may be added to the drawn fibers obtained in the drawing step by a crimper or the like. The number of crimps added in the crimping step is not particularly limited, but is preferably 10 to 25 crests/2.54 cm, for example, and can be adjusted by appropriately changing the stuffing box pressure in a pressing type crimper.

(Heat treatment step)

[0049] The drawn fibers obtained in the drawing step may be heat-treated. A heat treatment after drawing increases the crystallinity of the polyester resin, which is the first component of the heat-fusible composite fiber, which makes it possible to improve the bulkiness of non-woven fabrics. The heat treatment temperature is not particularly limited, but the heat treatment is preferably performed at a temperature range higher than 30°C to 70°C, which is the glass transition temperature of the polyester resin, and lower than the melting point of the polyethylene resin.

(Cutting step)

[0050] In a case where a carding step is employed when processing into non-woven fabrics using the composite fiber of the present invention, the composite fiber is required to be cut to an arbitrary length to pass through a carding machine. The length to which the composite fiber is cut, that is, the cut length, is preferably 30 to 64 mm from the viewpoint of fineness and the passing performance of the carding machine.

(Fiber treatment agent adding step)

[0051] The surface of the composite fiber of the present invention may be treated with various fiber treatment agents, which makes it possible to impart functions such as hydrophilicity, water repellency, antistatic properties, surface smoothness, and abrasion resistance.

[0052] Examples of the fiber treatment agent adhesion step include a method of adhering a fiber treatment agent with a kiss roll when taking up undrawn fibers, and a method of adhering by a touch roll method, an immersion method, a spraying method, or the like during drawing and/or after drawing.

(Non-woven fabric)

[0053] A non-woven fabric of the present invention contains the heat-fusible composite fibers described above, and thus reduces the consumption of fossil resources and is excellent in bulkiness and flexibility.

[0054] The biomass-derived carbon content of the non-woven fabric in the present invention is not particularly limited, but is preferably 10% or more, more preferably 15% to 60%, and further preferably 25% to 40% or more from the viewpoint of reducing the consumption of fossil resources. In order to obtain such a non-woven fabric having a biomass-derived carbon content of 10% or more, only composite fibers having a biomass-derived carbon content of 10% or more may be used, or a total biomass-derived carbon content may be set to 10% or more by mixing with other fibers. Examples of the other fibers include natural fibers (such as wood fibers), regenerated fibers (such as rayon), semi-synthetic fibers (such as acetate), chemical fibers, and synthetic fibers (such as polyester, acrylic, nylon, and vinyl chloride). As long as the effects of the present invention are not impaired, the mixing ratio of such fibers other than the heat-fusible composite fibers is not limited, but can be 1% to 50% by weight, for example.

[0055] The basis weight of the non-woven fabric is not particularly limited, but is preferably 15 to 40 g/m² and more preferably 18 to 30 g/m², especially when used as a non-woven fabric for sanitary materials. When the basis weight is 15 g/m² or more, this is preferable because then texture and cushioning properties are maintained, which makes it possible to prevent liquid return, and when the basis weight is 40 g/m² or less, this is preferable because then surface smoothness, air breathability, and liquid permeability can be maintained.

[0056] The specific volume of the non-woven fabric is not particularly limited, but is preferably 30 to 100 cm³/g and is more preferably 50 to 70 cm³/g, especially when used as a non-woven fabric for sanitary materials. The specific volume is a parameter used as an index of bulkiness, and as the specific volume becomes larger, the non-woven fabric can be evaluated to be bulkier. When the specific volume is 30 cm³/g or more, a bulkiness that can be applied as sanitary materials can be obtained, and when the specific volume is 100 cm³/g or less, this is preferable because then the strength of the non-woven fabric increases, and the non-woven fabric does not become too thick, resulting in excellent workability into sanitary materials.

[0057] The strength in the longitudinal direction (MD strength) of the non-woven fabric is not particularly limited, but is preferably 35 N/50 mm or more, and more preferably 45 N/50 mm or more. When the MD strength of the non-woven fabric is 35 N/50 mm or more, this is preferable because this results in excellent workability into sanitary materials.

[0058] The non-woven fabric of the present invention may consist of one type of (single layer) non-woven fabric, or may be a laminate of two or more types of non-woven fabrics in which composite fibers used are different in fineness, composition, density, and the like. When two or more types of non-woven fabrics are laminated, for example, by laminating non-woven fabrics having composite fibers with different fineness, a non-woven fabric in which the size of the gap configured between the fibers changes in the thickness direction of the non-woven fabric is configured, which makes it possible to control liquid permeability, a liquid permeation speed, the texture of a surface layer, and the like. Furthermore, for example, by laminating non-woven fabrics having composite fibers with different compositions, a non-woven fabric in which the hydrophilicity and the hydrophobicity of the non-woven fabric change in the thickness direction of the non-woven fabric is formed, which makes it possible to control liquid permeability and a liquid permeation speed.

[0059] The non-woven fabric of the present invention is not particularly limited, but may be laminated and integrated with other through-air non-woven fabric, airlaid non-woven fabric, spunbond non-woven fabric, melt-blown non-woven fabric, spunlace non-woven fabric, needle-punched non-woven fabric, non-woven fabrics such as films, meshes, and nets; films; and sheets. By laminating and integrating, liquid permeability, a liquid permeation speed, liquid return, and the like can be controlled. A method of laminating and integrating is not particularly limited, but examples thereof include a method of laminating and integrating using an adhesive such as hot melt, and a method of laminating and integrating by thermal adhesion such as through-air or heat embossing.

[0060] Within a range not impairing the effects of the present invention, the non-woven fabric may be subjected to shaping processing, perforating processing, antistatic processing, water repellent processing, hydrophilic processing, antibacterial processing, ultraviolet absorption processing, near-infrared absorption processing, electret processing, or the like depending on the purpose.

(Method for producing non-woven fabric)

[0061] A method for producing the non-woven fabric is not particularly limited, and examples thereof include a method of forming a web containing the above-mentioned heat-fusible composite fibers to integrate by heat or entanglement.

[0062] A method of forming webs is not particularly limited, long fiber webs configured by a spunbond method, a melt-blown method, a tow opening method, or the like may be used, or short fiber webs configured using short fibers (staples and chops) by a carding method, an airlaid method, a wet method, or the like may be used, among which the carding method or the airlaid method is preferable and the carding method is more preferable from the viewpoint of imparting bulkiness and flexibility to the non-woven fabric. In the present invention, the term "web" refers to a fiber assembly in the state in which fibers are not a little entangled, and means the state in which intersection points of the heat-fusible composite fibers are not fused.

[0063] A method of integrating webs by heat or entanglement is not particularly limited, and examples thereof include a through-air method, a thermal calendering method, a hydroentanglement method, and a needle-punching method, among which the through-air method is preferable from the viewpoint of imparting bulkiness and flexibility to the non-woven fabric. As the through-air method, well-known pieces of equipment and conditions, such as a method of heat-fusing composite fibers by a heat treatment device (for example, a hot air current penetration type heat treatment machine, a hot air current blowing type heat treatment machine) which is equipped with a conveying support that supports and conveys webs, may be applied.

[0064] The heat-fusible composite fiber of the present invention can be used for applications to various fiber products, which reduce the consumption of fossil resources and require bulkiness and flexibility, such as sanitary materials such as diapers, napkins, and incontinence pads; medical supplies such as masks, gowns, and surgical gowns; interior materials such as wall sheets, shoji paper, and flooring; daily life-related materials such as fabric covers, wipers for cleaning, and plastic food waste bags; toiletries products such as disposable toilets and plastic toilet bags; pet supplies such as pet sheets, pet diapers, and pet towels; industrial materials such as wiping materials, filters, cushioning materials, oil adsorbing materials, and ink tank adsorbents; covering materials; poultice bags; bedding materials; and nursing care products.

[Examples]

[0065] The present invention will be described in more detail below with reference to examples, but the scope of the present invention is not limited to these.

[0066] Evaluation of physical properties in the present invention was perconfigured by the following method.

<Biomass-derived carbon content>

[0067] The total carbon and ^{14}C contents of a sample was measured using an accelerator mass spectrometer (AMS) (in which tandem accelerator and mass spectrometer were combined). From the total carbon and ^{14}C contents in the

sample, the biomass-derived carbon content of the carbon contained in the sample was calculated according to the following formula.

$$\text{Biomass-derived carbon content (\%)} = (\text{biomass-derived carbon (}^{14}\text{C) amount in sample} / \text{total carbon amount in sample}) \times 100$$

<Intrinsic viscosity of polyester resin>

[0068] The measurement was perconfigured according to JIS K 7367-1.

<MFR of polyethylene resin>

[0069] The melt mass flow rate (MFR) was measured according to JIS K 7210. The measurement was perconfigured according to a condition D (test temperature: 190°C, load: 2.16 kg) in Table 1 of Annex A.

<Fineness of undrawn fibers, fineness of heat-fusible composite fibers, breaking strength, breaking elongation>

[0070] The measurement was perconfigured according to JIS-L-1015.

<Heat of fusion of polyester resin in heat-fusible composite fibers>

[0071] Using a differential scanning calorimetric measurement device (DSC8500) manufactured by PerkinElmer Japan G.K., the heat of fusion of the polyester resin in the composite fibers was measured according to the following procedure. First, the composite fibers were cut such that the mass was 4.20 to 4.80 mg, and a sample pan was filled therewith and covered. Then, measurement was perconfigured from 30°C to 300°C at a temperature rising rate of 10°C/minute in N₂ purge to obtain a melting chart. The obtained chart was analyzed to calculate the heat of fusion of the polyester resin from the area of the endothermic peak in the range of 245°C to 250°C.

<Basis weight of non-woven fabric>

[0072] Three pieces of non-woven fabric were cut out into squares of 10 cm × 10 cm. The weight of each of them was measured to be converted to a unit area, and the average value of the obtained values was taken as the basis weight of the non-woven fabric.

<Bulkiness of non-woven fabric>

[0073] Using a Digi-Thickness Tester manufactured by Toyo Seiki Seisaku-sho, Ltd., a pressure of 3.5 g/cm² was applied with a pressure element (load) having a diameter of 35 mm to measure the thickness at that time. The specific volume was calculated from the measured thickness using the following formula.

$$\text{Specific volume (cm}^3\text{/g)} = \text{thickness (mm)} / \text{basis weight (g/m}^2\text{)} \times 1,000$$

<MD strength of non-woven fabric>

[0074] A maximum strength when pulling a sample with a size of 50 mm × 150 mm and cut long in the longitudinal direction at a chuck distance of 100 mm and a tensile rate of 100 mm/minute using an Autograph (AGX-J) manufactured by Shimadzu Corporation was taken as the MD strength of the non-woven fabric.

flexibility of non-woven fabric>

[0075] A piece of non-woven fabric of 150 mm × 150 mm was cut out to perform a sensory test ("favorable" or "poor") by five panelists in terms of surface smoothness, cushioning properties, and draping properties, and the flexibility of the non-woven fabric was determined in the following three stages.

◎: "favorable" from all five panelists, which can be determined to have excellent flexibility.

O: "poor" from one panelist, which can be determined to have satisfactory flexibility.

Δ: "poor" from two to three panelists, which can be determined to have slightly inferior flexibility.

X: "poor" from four or more panelists, which can be determined to have poor flexibility.

5 **[0076]** The thermoplastic resins used in examples and comparative examples are as follows.

<Thermoplastic resin 1>

10 **[0077]** Biomass-derived polyethylene terephthalate (abbreviation: bio-PET) having an intrinsic viscosity of 0.65, a glass transition point of 70°C, and a biomass-derived carbon content of 30%.

<Thermoplastic resin 2>

15 **[0078]** Fossil-resource-derived polyethylene terephthalate (abbreviation: fossil PET) having an intrinsic viscosity of 0.64, a glass transition point of 70°C, and a biomass-derived carbon content of 0%.

<Thermoplastic resin 3>

20 **[0079]** Biomass-derived high-density polyethylene (abbreviation: bio-PE) having a density of 0.96 g/cm³, a MFR of 20 g/10 minutes, a melting point of 130°C, and a biomass-derived carbon content of 94%.

<Thermoplastic resin 4>

25 **[0080]** Fossil-resource-derived high-density polyethylene (abbreviation: fossil PE) having a density of 0.96 g/cm³, a MFR of 16 g/10 minutes, a melting point of 130°C, and a biomass-derived carbon content of 0%.

[Examples 1 to 6 and Comparative Examples 1 to 4]

30 **[0081]** Heat-fusible composite fibers and non-woven fabrics of examples and comparative examples were produced according to the conditions shown in Tables 1 and 2.

(Production of heat-fusible composite fiber)

35 **[0082]** Using the resins shown in Tables 1 and 2, spinning was perconfigured at a spinning temperature of 305°C with the first component/second component ratio (weight ratio) shown in Table 1, thereby obtaining undrawn fibers having a concentric sheath-core structure in which the first component was allocated on a core side and the second component was allocated on a sheath side.

40 **[0083]** The obtained undrawn fibers were subjected to a drawing step under the conditions shown in Tables 1 and 2 using a drawing machine. Thereafter, crimping was perconfigured such that the number of crimps was 16 crests/2.54 cm, a heat treatment was perconfigured for 5 minutes at the heat treatment temperature shown in Table 1, and cutting was perconfigured such that the fiber length was 44 mm, thereby obtaining heat-fusible composite fibers.

(Non-woven fabric processing)

45 **[0084]** The obtained heat-fusible composite fibers were passed through a roller carding machine to collect a fiber web. A piece of 100 cm × 30 cm was cut out from the fiber web and heat-treated at a processing temperature of 130°C using a hot air current circulation type heat treatment machine to heat-fuse the sheath component, thereby obtaining a non-woven fabric.

50 **[0085]** Tables 1 and 2 collectively show the production conditions and the physical properties evaluation results of each example and comparative example.

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[Table 1]

			Example 1	Example 2	Example 3	Example 4	Example 5	Example 6
First component	Mixing ratio of bio-PET	% bv weight	25	0	50	0	100	25
	Mixing ratio of fossil PET	% by weight	75	100	50	100	0	75
	Biomass- derived carbon content	%	7.5	0.0	150	0.0	30.0	7.5
Second component	Mixing ratio of bio-PE	% by weight	50	70	45	85	25	50
	Mixing ratio of fossil PE	% by weight	50	30	55	15	75	50
	Biomass- derived carbon content	%	47.0	65.8	42.3	79.9	23.5	47.0
	MFR	g/10 minutes	18	19	18	20	17	18
First component/second component		Weight/ weight	50/50	60/40	60/40	60/40	50/50	50/50
Fineness of undrawn fiber		dtex	8	8	8	8	8	10
Draw ratio		Ratio	5.7	5.5	5.5	5.6	5.6	4
Draw temperature		°C	105	105	105	105	105	105
Draw rate		m/ minute	50	50	50	50	50	50
Heat treatment temperature		°C	120	120	120	120	120	120
Composite fiber	Fineness	dtex	1.8	1.9	1.6	2.2	2.2	26
	Breaking strength	cN/dtex	2.3	2.3	2.3	2.0	2.2	2.4
	Breaking elongation	%	65	116	92	60	68	170
	Heat of fusion of polyester resin	J/g	26.0	29.9	29.7	29.9	25.0	24.2
	Biomass- derived carbon content	%	27.3	26.3	25.9	32.0	26.8	27.3

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(continued)

			Example 1	Example 2	Example 3	Example 4	Example 5	Example 6
Non-woven fabric	Basis weight	g/m ²	28	24	25	26	25	25
	Specific volume	cm ³ /g	62	66	60	63	62	70
	MD strength	N/50 mm	72	46	40	47	45	65
	Flexibility	-	☉	☉	☉	○	○	Δ
	Biomass-derived carbon content	%	27.3	26.3	25.9	32.0	26.8	27.3

[Table 2]

			Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4
First component	Mixing ratio of bio-PET	% by weight	0	0	50	0
	Mixing ratio of fossil PET	% by weight	100	100	50	100
	Biomass-derived carbon content	%	0.0	0.0	15.0	0.0
Second component	Mixing ratio of bio-PE	% bv weight	100	100	15	0
	Mixing ratio of fossil PE	% bv weight	0	0	85	100
	Biomass-derived carbon content	%	94.0	94.0	14.1	0.0
	MFR	g/10 minutes	20	20	16	16
First component/Second component		Weight/weight	50/50	50/50	60/40	50/50
Fineness of undrawn fiber		dtex	10	10	10	10
Draw ratio		Ratio	7.1	4.4	4.6	6.5
Draw temperature		°C	105	90	105	105
Draw rate		m/minute	50	85	50	50
Heat treatment temperature		°C	115	120	115	115

(continued)

			Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4
Composite fiber	Fineness	dtex	2.2	3.1	2.3	1.7
	Breaking strength	cN/dtex	1.3	2.2	2.1	2.3
	Breaking elongation	%	136	28	70	80
	Heat of fusion of polyester resin	J/g	22.4	24.6	23.3	23.5
	Biomass- derived carbon content	%	47.0	47.0	14.6	0.0
Non-woven fabric	Basis weight	g/m ²	23	25	25	25
	Specific volume	cm ³ /g	26	88	45	55
	MD strength	N/50 mm	29	45	42	40
	Flexibility	-	X	X	Δ	Δ
	Biomass- derived carbon content	%	47.0	47.0	14.6	0.0

[0086] From the results in Tables 1 and 2, in Examples 1 to 6 according to the present invention, the mixing ratio of the biomass-derived polyethylene resin and the fossil-resource-derived polyethylene resin as the polyethylene resin was 20:80 to 90:10. With such heat-fusible composite fibers, the biomass-derived carbon content was high, and even with a small fineness, the non-woven fabric maintained its bulkiness and had satisfactory flexibility. Particularly in Examples 1 to 3, the composite fibers had a small fineness and were very excellent in flexibility.

[0087] On the other hand, the composite fibers of Comparative Example 1 had a high mixing ratio of the biomass-derived polyethylene resin and a small specific volume (low bulk). This is thought to be caused because the high mixing ratio of the biomass-derived polyethylene resin made the molecular weight decrease significant during the melting process of the resin, and thereby sufficient drawability was not obtained, resulting in a decrease in the degree of crystallinity of the polyester resin. Furthermore, when the draw temperature was lowered to increase the specific volume (increase the bulk), the fineness increased, resulting in impairing of flexibility (Comparative Example 2). Furthermore, the composite fibers of Comparative Example 3 had a low mixing ratio of the biomass-derived polyethylene resin, resulting in slightly inferior bulkiness and flexibility, which made them generally difficult to be applied as sanitary materials. In Comparative Example 4 which did not contain the biomass-derived resin, an acceptable bulkiness was obtained. However, not only flexibility was slightly inferior, but also the biomass-derived carbon content was low, and thereby the consumption of fossil resources could not be reduced.

[Industrial Applicability]

[0088] In the heat-fusible composite fiber of the present invention, by mixing the biomass-derived polyethylene resin and the fossil-resource-derived polyethylene resin in an appropriate ratio as the polyethylene resin forming the second component, it is possible to provide the non-woven fabric that reduces the consumption of fossil resources and has excellent bulkiness and flexibility. Therefore, the heat-fusible composite fiber of the present invention can be used for applications to various fiber products, which reduce the consumption of fossil resources and require bulkiness and

flexibility, such as sanitary materials such as diapers, napkins, and incontinence pads; medical supplies such as masks, gowns, and surgical gowns; interior materials such as wall sheets, shoji paper, and flooring; daily life-related materials such as fabric covers, wipers for cleaning, and plastic food waste bags; toiletries products such as disposable toilets and plastic toilet bags; pet supplies such as pet sheets, pet diapers, and pet towels; industrial materials such as wiping materials, filters, cushioning materials, oil adsorbing materials, and ink tank adsorbents; covering materials; poultice bags; bedding materials; and nursing care products.

Claims

1. A heat-fusible composite fiber comprising:

a first component configured from a polyester resin; and
a second component configured from a polyethylene resin having a lower melting point than that of the first component,
wherein a mixing ratio (weight ratio) of a biomass-derived polyethylene resin and a fossil-resource-derived polyethylene resin in the polyethylene resin is 20:80 to 90:10.

2. The heat-fusible composite fiber according to claim 1, wherein a biomass-derived carbon content in the polyethylene resin is 20% to 90%.

3. The heat-fusible composite fiber according to claim 1 or 2, wherein a biomass-derived carbon content in the heat-fusible composite fiber is 10% or more.

4. The heat-fusible composite fiber according to any one of claims 1 to 3, wherein a biomass-derived carbon content in the polyester resin is 30% or less.

5. The heat-fusible composite fiber according to any one of claims 1 to 4, wherein a fineness of the heat-fusible composite fiber is 2.2 dtex or less.

6. The heat-fusible composite fiber according to any one of claims 1 to 5, wherein a heat of fusion of the polyester resin in the heat-fusible composite fiber is 24 J/g or more.

7. The heat-fusible composite fiber according to any one of claims 1 to 6,
wherein the heat-fusible composite fiber is a sheath-core type heat-fusible composite fiber comprising:

the first component as a core component; and
the second component as a sheath component.

8. The heat-fusible composite fiber according to any one of claims 1 to 7, wherein the polyester resin is polyethylene terephthalate, and the polyethylene resin is high-density polyethylene.

9. A non-woven fabric, comprising the heat-fusible composite fiber according to any one of claims 1 to 8.

10. An absorbent article, using the heat-fusible composite fiber according to any one of claims 1 to 8.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2022/005032

A. CLASSIFICATION OF SUBJECT MATTER

D01F 8/06(2006.01)i; **D01F 8/14**(2006.01)i; **D04H 1/541**(2012.01)i
FI: D01F8/06; D01F8/14 Z; D04H1/541

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

D01F8/00-8/18; D04H1/00-18/04; A61F13/00-13/84; A61L15/00-15/64

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
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)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2010-65342 A (NIPPON ESTER CO., LTD.) 25 March 2010 (2010-03-25) example 3, paragraphs [0027], [0030]	1-3, 5-7, 9-10
Y	example 3, paragraphs [0027], [0030]	1-10
Y	JP 2018-199885 A (TORAY INDUSTRIES, INC.) 20 December 2018 (2018-12-20) claims, paragraphs [0082], [0089], examples	1-10
A	JP 2011-038207 A (UNITIKA LTD.) 24 February 2011 (2011-02-24)	1-10

☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

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“Y” 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

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Japan Patent Office (ISA/JP)
3-4-3 Kasumigaseki, Chiyoda-ku, Tokyo 100-8915
Japan

Authorized officer

Telephone No.

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INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.

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Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
JP 2010-65342 A	25 March 2010	(Family: none)	
JP 2018-199885 A	20 December 2018	(Family: none)	
JP 2011-038207 A	24 February 2011	(Family: none)	

Form PCT/ISA/210 (patent family annex) (January 2015)

REFERENCES CITED IN THE DESCRIPTION

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