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(54) **RANDOM MAT AND PRODUCTION METHOD THEREFOR, AND FIBER-REINFORCED RESIN MOLDED MATERIAL USING RANDOM MAT**

WIRRVLIIES UND HERSTELLUNGSVERFAHREN DAFÜR SOWIE FASERVERSTÄRKTES HARZFORMMATERIAL UNTER VERWENDUNG EINES WIRRVLIESES

MAT ALÉATOIRE ET SON PROCÉDÉ DE PRODUCTION, ET MATÉRIAU MOULÉ EN RÉSINE RENFORCÉ PAR DES FIBRES METTANT EN OEUVRE LE MAT ALÉATOIRE

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**Description**Technical Field of the Invention

5 **[0001]** The present invention relates to a random mat comprising a chopped fiber bundle of reinforcing fibers, a method for producing the random mat, and a fiber-reinforced resin molding material using the random mat.

Background Art of the Invention

10 **[0002]** Known is a technology for molding a molded article having a desired shape by heating and pressurizing molding, using a fiber-reinforced resin molding material comprising a random mat comprising chopped fiber bundles which is randomly dispersed with fiber bundles of discontinuous reinforcing fibers (hereinafter, referred to as chopped fiber bundles) prepared by cutting a fiber bundle of continuous reinforcing fibers (for example, carbon fibers) and a matrix resin (for example, a thermosetting resin or a thermoplastic resin) (for example, Patent documents 1, 2). In such a  
15 conventional fiber-reinforced resin molding material, in case where the fiber bundle in the fiber-reinforced resin molding material is composed of a fiber bundle formed from a predetermined strand and having a predetermined number of single fibers, usually, in a molding material comprising a fiber bundle with a large number of single fibers, although the flowability during molding is excellent, the mechanical properties of a molded article tend to be inferior.

20 **[0003]** For example, in Patent document 1, a molding material, in which the number of filaments of a chopped fiber bundle in the molding material is specified within a range of 10,000 to 700,000, is disclosed. In such a molding material, since the number of filaments of the fiber bundle is large, at the time of molding, reinforcing fibers can move efficiently in a form of a fiber bundle together with a resin, so that an excellent flowability can be obtained, but with respect to a molded article after molding with this molding material, there is a high possibility that stress concentration occurs at the end portion of the fiber bundle and the like in the molded article, for example when the molded article breaks, and  
25 therefore, it is not suitable for molding a molded article required with high mechanical properties.

**[0004]** On the other hand, for example, in Patent document 2, a fiber-reinforced resin using a fiber bundle which is separated so that the number of single fibers becomes 100 or less is disclosed, but as compared with the embodiment disclosed in the above-described Patent document 1, since the number of single fibers of the fiber bundle is much smaller, the reinforcing fibers are well dispersed in a molded article, the possibility of stress concentration occurring at  
30 the end portion of the fiber bundle or the like in the molded article decreases, and the mechanical properties of the molded article are increased, on the other hand, a possibility is left that high flowability cannot be obtained at the time of molding as expected.

**[0005]** Thus, in a fiber-reinforced resin molding material using a fiber bundle having a relatively large number of single fibers, excellent productivity and excellent flowability during molding tend to be obtained, but the mechanical properties of a molded article tend to be inferior, and on the contrary, in a fiber-reinforced resin molding material using a fiber bundle having a relatively small number of single fibers, the mechanical properties of a molded article tend to be excellent, but it tends to be difficult to improve the flowability during molding.

**[0006]** Focusing on such a tendency, in Patent document 3, a fiber-reinforced resin molding material comprising at least bundled aggregates of discontinuous reinforcing fibers and a matrix resin is proposed wherein the bundled aggregate of the reinforcing fibers contains both of reinforcing fiber aggregates A formed by cutting a strand of continuous reinforcing fibers after subjecting the strand to a splitting treatment to completely divide the strand into a plurality of bundles, and reinforcing fiber aggregates B containing undivided portions which are not subjected to the splitting treatment or/and in which the splitting treatment is insufficient, at a predetermined proportion. This proposal makes it possible to achieve both of good flowability during molding and excellent mechanical properties of a molded article at a good balance.  
40 However, in the fiber-reinforced resin molding material described in Patent document 3, because basically the bundled aggregate of the discontinuous reinforcing fibers was formed by being cut in the direction perpendicular to the lengthwise direction of the fiber bundle, there is a possibility that a tendency is left in that a stress concentration is liable to occur at the end portion of the bundled aggregate in a molded article.

**[0007]** On the other hand, as another manner for achieving both of good flowability during molding and excellent mechanical properties of a molded article at a good balance, in Patent document 4, described is a chopped fiber bundle in which transition sections each in which the number of reinforcing fibers changes toward each tip of the fiber bundle are provided at both end portions of the fiber bundle, and in which the change amount per unit length of the total cross-sectional area of reinforcing fibers in the cross section of the fiber bundle is suppressed to be small between both ends. However, since the chopped fiber bundle described in Patent document 4 is obtained basically by cutting a continuous reinforcing fiber bundle in which a predetermined number of continuous reinforcing fibers are bundled, in particular, in  
55 such a case where the number of single fibers of the chopped fiber bundle is great and the fiber bundle is thick, there is also a possibility that a tendency is left in that a stress concentration is liable to occur at the end portion of the chopped fiber bundle in a molded article.

**[0008]** Patent Document 5 discloses a random mat comprising at least one chopped fiber bundle satisfying the requirements (b), (c) and (d) of present claim 1. Cutting of the bundle is performed at an oblique angle.

**[0009]** Thus, according to the proposals of Patent documents 3-5, improvements are being promoted to achieve both of good flowability during molding and excellent mechanical properties of a molded article at a good balance, but better flowability during molding, higher mechanical properties (strength, modulus of elasticity) of a molded article and further reduction of its variation are being required than those in these proposals.

Prior art documents

Patent documents

**[0010]**

Patent document 1: JP-A-2013-202890

Patent document 2: JP-A-2008-174605

Patent document 3: WO2016 / 043037

Patent document 4: Japanese Patent No. 5672947

Patent document 5: EP 2 151 418 A1

Summary of the Invention

Problems to be solved by the Invention

**[0011]** Accordingly, in view of the requirements as described above, an object of the present invention is to provide a random mat capable of achieving better flowability during molding, higher mechanical properties (strength, modulus of elasticity) of a molded article and further reduction of its variation as compared with the conventional technologies, a method for producing the same, and a fiber-reinforced resin molding material using the same.

Means for solving the Problems

**[0012]** To achieve the above-described object, a random mat according to the present invention comprises at least a chopped fiber bundle [A], and the chopped fiber bundle [A] satisfies at least the following (a) to (d):

(a) the chopped fiber bundle [A] is a bundled aggregate of discontinuous reinforcing fibers obtained by cutting a partially separated fiber bundle [B] prepared by alternately forming separation-processed sections, each of which is separated into a plurality of bundles, and not-separation-processed sections, along a lengthwise direction of a fiber bundle comprising a plurality of reinforcing fibers;

(b) the chopped fiber bundle [A] has a first transition section in which the number of the reinforcing fibers in a cross section of the fiber bundle in a direction perpendicular to the lengthwise direction of the fiber bundle is increased from a first tip which is one tip in the lengthwise direction of the fiber bundle, toward a second tip which is the other tip, and a second transition section in which the number of the reinforcing fibers in the cross section of the fiber bundle is increased from the second tip toward the first tip;

(c) between the first transition section and the second transition section, along the lengthwise direction of the fiber bundle, an unchanging section in which the number of the reinforcing fibers in the cross section of the fiber bundle is invariable is provided, and one end surface of the unchanging section coincides with a first terminal end surface which is a terminal end at a side opposite to the first tip of the first transition section and the other end surface of the unchanging section coincides with a second terminal end surface which is a terminal end at a side opposite to the second tip of the second transition section, or the first terminal end surface and the second terminal end surface coincide directly with each other; and

(d) a change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip and the second tip is  $0.05\text{mm}^2$  or less per a length of 1 mm in the lengthwise direction of the fiber bundle, wherein the partially separated fiber bundle [B] is cut at an angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) with respect to the lengthwise direction of the fiber bundle, when obtaining the chopped fiber bundle [A].

**[0013]** In such a random mat according to the present invention, the chopped fiber bundle [A] is a bundled aggregate of discontinuous reinforcing fibers which is obtained by cutting a partially separated fiber bundle [B] prepared by alternately forming separation-processed sections, each of which is separated into a plurality of bundles, and not-separation-processed sections, along a lengthwise direction of a fiber bundle comprising a plurality of reinforcing fibers, and which

has a first transition section and a second transition section, each in which the number of the reinforcing fibers in a cross section of the fiber bundle in a direction perpendicular to the lengthwise direction of the fiber bundle is changed, in both end portion sides in the lengthwise direction of the fiber bundle. Namely, although in the fiber-reinforced resin molding material in Patent document 3, the bundled aggregate [A] of discontinuous reinforcing fibers was formed by being cut in the direction perpendicular to the lengthwise direction of the fiber bundle, in the present invention, in particular, a partially separated fiber bundle is obliquely cut to the lengthwise direction of the fiber bundle so as to have the first transition section and the second transition section. By being obliquely cut relative to the lengthwise direction of the fiber bundle, it becomes possible for the cutting plane to extend over the separation-processed section and the not-separated-processed section, whereby in particular the end portion of the formed bundled aggregate [A] is easily formed in a shape in which stress does not easily concentrate in a molded article (various examples will be described later), and further, it becomes also possible to make the fiber bundle like the reinforcing fiber aggregate B in Patent document 3 smaller in width. Further, since the fiber bundle used for forming the chopped fiber bundle [A] is the partially separated fiber bundle [B] in which the separation-processed sections and the not-separation-processed sections are alternately formed, as compared with the chopped fiber bundle obtained by cutting a continuous reinforcing fiber bundle in which a predetermined number of continuous reinforcing fibers are bundles such as one in Patent document 4, stress concentration is less likely to occur at the end portion of the chopped fiber bundle in a molded article. Furthermore, since between the first tip and the second tip of the chopped fiber bundle, the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip and the second tip is suppressed to be small at 0.05mm<sup>2</sup> or less per a length of 1 mm in the lengthwise direction of the fiber bundle, the stress concentration at the end portion of the chopped fiber bundle in a molded article can be prevented more effectively and smoothly. Namely, the state where the number of reinforcing fibers increases from a tip to an end of the transition section of the chopped fiber bundle [A] in the present invention can be expressed conversely as a state where the number of reinforcing fibers decreases from the central portion of the chopped fiber bundle [A] toward a tip of the chopped fiber bundle [A]. By this state decreased with the number of reinforcing fibers, the occurrence of stress concentration in a molded article can be prevented. This state decreased with the number of reinforcing fibers is preferably a formation in which the number of reinforcing fibers decreases gradually, that is, continuously. If chopped fiber bundle [A] is thick, the number of reinforcing fibers is large, and the total cross-sectional area of reinforcing fibers is larger, the effect of preventing the occurrence of stress concentration will be greater. The larger the total cross-sectional area of the reinforcing fibers, the larger the load born by one chopped fiber bundle [A] in a molded article becomes, but even if the load to be born is large, the state where the load is delivered at a stroke to the end portion of an adjacent chopped fiber bundle [A] through a matrix resin can be effectively prevented by the state decreased with the number of reinforcing fibers in the transition section. Namely, the transmission of the load between adjacent chopped fiber bundles [A] is gradually performed due to the state where the number of reinforcing fibers in the transition section is decreased, in particular, the state without a sudden change in the number, the stress concentration at the end portion of the chopped fiber bundle [A] is more effectively prevented, and the occurrence of stress concentration over the entire chopped fiber bundle [A] is more effectively prevented. As a result, it becomes possible to exhibit higher mechanical properties (strength, elastic modulus) and to further reduce variations in the mechanical properties in a molded article. With respect to good flowability during molding, it is ensured by a condition where the partially separated fiber bundle is cut into a chopped fiber bundle [A] which is a bundled aggregate of discontinuous reinforcing fibers.

**[0014]** Where, in the above-described random mat according to the present invention, it is sufficient that the chopped fiber bundle [A] satisfying the above-described (a) to (d) is contained, and a chopped fiber bundle not satisfying all of (a) to (d) (for example, a chopped fiber bundle in which the above-described (a) is satisfied, and in the above-described (d), "between the first tip and the second tip, the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle exceeds 0.05 mm<sup>2</sup> per a length of 1 mm in the lengthwise direction of the fiber bundle, and the above-described (b) and (c) are not satisfied, or are satisfied) may be contained in a range which does not damage the object and effect of the present invention.

**[0015]** In the above-described random mat according to the present invention, a formation may be employed wherein in the partially separated fiber bundle [B], an entangled part in which the reinforcing fibers are entangled at least at one end of at least one of the separation-processed sections, and/or an entanglement accumulation part in which the entangled parts are accumulated, is formed.

**[0016]** Further, in the random mat according to the present invention, a formation may be employed wherein bundled aggregate of the discontinuous reinforcing fibers obtained by cutting the partially separated fiber bundle [B] are classified into at least the following aggregates [X] to [Z], and the chopped fiber bundle [A] comprises at least one of the aggregates [X], [Y] and [Z]:

aggregate [X]: a separated bundle aggregate divided into arbitrary number of bundles by separation processing;  
aggregate [Y]: a joined bundle aggregate in which the reinforcing fibers of fiber bundles are joined to each other by being formed with the not-separation-processed section, and/or an entangled part in which the reinforcing fibers

are entangled at least at one end of at least one of the separation-processed sections, and/or an entanglement accumulation part in which the entangled parts are accumulated; and aggregate [Z]: a joined cut aggregate in which a cutting plane at cutting of the partially separated fiber bundle is intersected with the not-separation-processed section and/or the entangled part and/or the entanglement accumulation part, and at an intersected part, the joining of the reinforcing fibers of fiber bundles to each other is cut.

**[0017]** In this formation, it is preferred that the content of the joined bundle aggregate [Y] in the bundled aggregates of the discontinuous reinforcing fibers obtained by cutting the partially separated fiber bundle [B] is in the range of 0 to 15%. Namely, the joined bundle aggregate [Y] may not be contained, but in case of being contained, it is preferred to suppress the content at 15% at highest.

**[0018]** The present invention also provides a method for producing a random mat as described above. Namely, a method for producing a random mat according to the present invention is a method for producing a random mat as described above. wherein the partially separated fiber bundle [B] is cut at an angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) with respect to the lengthwise direction of the fiber bundle when obtaining the chopped fiber bundle [A].

**[0019]** In this method for producing a random mat, it is preferred that the partially separated fiber bundle [B] is cut so as to satisfy the following equation (1) when obtaining the chopped fiber bundle [A].

$$W \cdot \cos \theta / D \geq 3 \cdots (1)$$

W: width of fiber bundle when cutting partially separated fiber bundle

D: distance between cutting planes in chopped fiber bundle [A]

**[0020]** Furthermore, the present invention also provides a fiber-reinforced resin molding material comprising the above-described random mat and a matrix resin [M].

#### Effect according to the Invention

**[0021]** According to the random mat and its production method and the fiber-reinforced resin molding material using the same according to the present invention, by having the specific chopped fiber bundle [A] of the discontinuous reinforcing fibers, which is formed by cutting the partially separated fiber bundle [B] prepared by alternately forming separation-processed sections and not-separation-processed sections, obliquely with respect to the lengthwise direction of the fiber bundle, excellent flowability during molding can be realized, extremely high mechanical properties (strength, elastic modulus) when made into a molded article can be realized, and variations in the mechanical properties can be suppressed to be small.

#### Brief explanation of the drawings

##### **[0022]**

[Fig. 1] Fig. 1 is a schematic perspective view showing a partially separated fiber bundle [B] and a cutting example thereof in the present invention.

[Fig. 2] Fig. 2 is a schematic plan view of a fiber bundle showing an embodiment of a partially separated fiber bundle [B] in the present invention.

[Fig. 3] Fig. 3 is a schematic plan view of a fiber bundle showing another embodiment of a partially separated fiber bundle [B] in the present invention.

[Fig. 4] Fig. 4 is a schematic plan view of a fiber bundle showing a further embodiment of a partially separated fiber bundle in the present invention.

[Fig. 5] Fig. 5 shows a schematic plan view (A) and a schematic side view (B) showing an example of a method of manufacturing a partially separated fiber bundle [B] in the present invention.

[Fig. 6] Fig. 6 is a schematic plan view of a partially separated fiber bundle [B] showing a basic technical concept of oblique cutting in the present invention.

[Fig. 7] Fig. 7 is a schematic plan view of a partially separated fiber bundle [B] showing an example of orthogonal cutting.

[Fig. 8] Fig. 8 is a schematic plan view showing an example of a method for manufacturing a separated bundle aggregate [X] in the present invention.

[Fig. 9] Fig. 9 is a schematic plan view showing an example of a method for manufacturing a joined bundle aggregate [Y] in the present invention.

[Fig. 10] Fig. 10 is a schematic plan view showing another example of a method for manufacturing a joined bundle aggregate [Y] in the present invention.

[Fig. 11] Fig. 11 is a schematic plan view showing a further example of a method for manufacturing joined bundle aggregate [Y] in the present invention.

[Fig. 12] Fig. 12 is a schematic plan view showing an example of a method for manufacturing a joined cut aggregate [Z] in the present invention.

[Fig. 13] Fig. 13 is a schematic plan view for explaining equation (1) in the present invention.

[Fig. 14] Fig. 14 is a schematic plan view showing an example of a chopped fiber bundle [A] in the present invention.

[Fig. 15] Fig. 15 is a schematic plan view showing another example of a chopped fiber bundle [A] in the present invention.

[Fig. 16] Fig. 16 is a schematic plan view showing still another example of the chopped fiber bundle [A] in the present invention.

[Fig. 17] Fig. 17 is a schematic plan view showing further various examples of a chopped fiber bundle [A] in the present invention.

[Fig. 18] Fig. 18 shows a plan view (a), a side view (b) and a graph (c) showing a state of increase/decrease of the number of reinforcing fibers in the arrangement direction of reinforcing fibers of a chopped fiber bundle, of the chopped fiber bundle [A] shown in Fig. 15.

[Fig. 19] Fig. 19 shows a plan view (a), a side view (b) and a graph (c) showing a state of increase/decrease of the number of reinforcing fibers in the arrangement direction of reinforcing fibers of a chopped fiber bundle, of the chopped fiber bundle [A] shown in Fig. 16.

[Fig. 20] Fig. 20 is a schematic plan view showing an example of a fiber-reinforced resin molding material of the present invention.

[Fig. 21] Fig. 21 is a schematic perspective view for explaining an example of a method for producing a fiber-reinforced resin molding material of the present invention.

#### Embodiments for carrying out the Invention

**[0023]** Hereinafter, the present invention will be explained in detail with reference to the drawings, together with embodiments.

**[0024]** First, the partially separated fiber bundle [B] in the present invention will be explained. First, referring to Fig. 1, a partially separated fiber bundle [B] 1, prepared by alternately forming separation-processed sections 2, each of which is separated into a plurality of bundles, and not-separation-processed sections 3, along a lengthwise direction L of a fiber bundle comprising a plurality of reinforcing fibers F, and its cutting, will be explained. As shown in Fig. 1, the partially separated fiber bundle [B] 1, prepared by alternately forming separation-processed sections 2 and not-separation-processed sections 3 along the lengthwise direction L of the fiber bundle, is run in the direction A, and the partially separated fiber bundle [B] 1 is cut by a cutting blade 4 in a direction traversing the fiber bundle to form a chopped fiber bundle [A] 5 comprising bundled aggregates of discontinuous reinforcing fibers. Here, as a preferable example of a method for obtaining the chopped fiber bundle [A] 5, exemplified is a method of cutting at an angle  $\theta$  with respect to the lengthwise direction of the fiber bundle. This cutting angle  $\theta$  is set at  $4^\circ \leq \theta \leq 25^\circ$ , to be a cutting in an oblique direction. Within such a range of cutting angle  $\theta$ , it becomes possible to realize good flowability during molding and high mechanical properties of a molded article and its low variation.

**[0025]** Although the above-described partially-separated fiber bundle [B] 1 before cutting basically has a form in which separation-processed sections 2 and not-separation-processed sections 3 as shown in Fig. 1 are alternately formed along the lengthwise direction of the fiber bundle, as shown in Fig. 2 or 3, an embodiment, wherein an entangled part 11 in which reinforcing fibers are entangled with each other and/or an entanglement accumulation part 12 in which the entangled parts are accumulated in at least one end portion of at least one separation-processed section 2, can be also employed.

**[0026]** Further, as shown in Fig. 4, a partially separated fiber bundle [B] 17 according to an embodiment, which comprises a combination of a formation in which the separation-processed sections 13 and the not-separation-processed sections 14 are alternately formed along the lengthwise direction of the fiber bundle and a formation in which the separation-processed sections 15 and the not-separation-processed sections 16 are alternately formed along the lengthwise direction of the fiber bundle, and wherein the separation-processed sections 15 of one formation are formed so as to extend over the not-separation-processed sections 14 of the other formation, is also included in the partially separated fiber bundle in the present invention.

**[0027]** The partially separated fiber bundle [B] in the present invention as described above is formed, for example, as shown in Fig. 5, although it is not particularly limited. Fig. 5 is a schematic plan view (A) and a schematic side view (B) showing an example in which a separation means 21 is pierced into a running fiber bundle 20. The running direction A of the fiber bundle (the arrow) in the figure is the lengthwise direction of the fiber bundle 20, indicating that the fiber

bundle 20 is continuously supplied from a fiber bundle supply device (not shown). The separation means 21 has a protrusion 22 having a protruding shape which is easy to be pierced into the fiber bundle 20, is pierced into the running fiber bundle 20, and generates separation-processed sections 23 substantially parallel to the lengthwise direction of the fiber bundle 20. Depending on the number of fiber bundles to be separated, it is also possible to simultaneously use a plurality of separation means 21. It is possible to arbitrarily arrange a plurality of protrusions 22 by a manner such as arranging a plurality of separation means 21 in parallel, by turns, or by shifting the phase.

**[0028]** In case where the fiber bundle 20 comprising a plurality of reinforcing fibers is divided into separated bundles having a lesser number of reinforcing fibers by the separation means 21, there is a possibility that, because a plurality of reinforcing fibers are not aligned substantially in the fiber bundle 20, but in single fiber level there are many portions where the fibers are entangled with each other, entangled parts 25, each in which reinforcing fibers are entangled with each other, may be formed in the vicinity of contact part 24 during the separation treatment. Here, "forming the entangled part 25" means, for example, a case of forming (moving) the entanglement of reinforcing fibers having existed beforehand in the separation-processed sections in the contact part 24 by the separation means 21, a case of forming (manufacturing) an aggregate newly entangled with reinforcing fibers by the separation means 21, or the like.

**[0029]** After creating separation-processed sections 23 in an arbitrary range, the separation means 21 is pulled out from the fiber bundle 20. By this pulling out, separation-processed sections 23 subjected to the separation processing are created, and at the same time entanglement accumulation parts 26 accumulated with entangled parts 25 are created. Further, fluffs generated from the fiber bundle during the separation processing may form a fluff accumulation 27 near the entanglement accumulation part 26 at the time of the separation processing.

**[0030]** Thereafter, by piercing the separation means 21 again into the fiber bundle 20, a not-separation-processed section 28 is created.

**[0031]** Next, in Fig. 6, the basic technical concept in the present invention employing the oblique cutting of a partially separated fiber bundle [B] will be explained as compared with a case of Fig. 7 employing the orthogonal cutting of a partially separated fiber bundle [B]. In Figs. 6 and 7, symbol 31 denotes a partially separated fiber bundle [B] wherein along the lengthwise direction of a fiber bundle comprising a plurality of reinforcing fibers, separation-processed sections 32 each of which is separated into a plurality of bundles and not-separation-processed sections 33 containing the aforementioned entangled part and the like are alternately formed. In Fig. 7, the cutting plane 35 for the partially-separated fiber bundle [B] 31 is set in a direction (90° direction) orthogonal to the lengthwise direction X-X of the fiber bundle, whereas in the present invention, the angle  $\theta$  of the cutting plane 34 with respect to the lengthwise direction X-X of the fiber bundle is set at an angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) in an oblique direction.

**[0032]** Then, when burning off a matrix resin [M] from a molded article molded by randomly dispersing a fiber-reinforced resin molding material containing chopped fiber bundles [A], which are bundled aggregates of discontinuous reinforcing fibers obtained by the above-described cutting, and the matrix resin [M], heating and pressing, and leaving only the bundled aggregates of discontinuous reinforcing fibers to observe them as a plan view, the distribution diagrams of discontinuous reinforcing fiber bundled aggregates, for example, as exemplified in the right sides of Fig. 6 and Fig. 7, are obtained. In the distribution diagram shown in Fig. 7, bundled aggregates 36, which are formed by cutting at both sides of not-separation-processed sections 33 containing mainly entangled parts etc. at cutting planes 35 and in which the end portions in the lengthwise direction of the fiber bundle are relatively wide and formed as end portions extending in the direction orthogonal to the lengthwise direction of the fiber bundle, are left substantially in forms similar to their original forms. In such end portions of bundled aggregates 36, as aforementioned, a stress concentration is liable to occur, and it causes a decrease in the mechanical properties of the molded article and its variation. On the other hand, in the distribution diagram shown in Fig. 6, there is no such bundled aggregate 36 in which stress concentration is liable to occur, and for example, even in bundled aggregates 37 formed by cutting obliquely including non-separation treatment sections 33 containing entangled parts etc., each bundled aggregate 37 has a relatively narrow width and becomes narrower as it goes to the end portion, and besides, it is formed in the form of a bundled aggregate which does not have an end portion where stress concentration is liable to occur as in the bundled aggregate 36. Therefore, it becomes possible to improve the mechanical properties of the molded article and to reduce the variation of the mechanical properties.

**[0033]** In the bundled aggregate of discontinuous reinforcing fibers obtained by cutting the partially separated fiber bundle [B] as described above is classified, for example, into at least the following aggregates [X] - [Z], the chopped fiber bundle [A] can be formed as a formation comprising at least one of the aggregates [X], [Y] and [Z]:

- aggregate [X]: a separated bundle aggregate divided into arbitrary number of bundles by separation processing;
- aggregate [Y]: a joined bundle aggregate in which the reinforcing fibers of fiber bundles are joined to each other by being formed with the not-separation-processed section, and/or an entangled part in which the reinforcing fibers are entangled at least at one end of at least one of the separation-processed sections, and/or an entanglement accumulation part in which the entangled parts are accumulated; and
- aggregate [Z]: a joined cut aggregate in which a cutting plane at cutting of the partially separated fiber bundle is

intersected with the not-separation-processed section and/or the entangled part and/or the entanglement accumulation part, and at an intersected part, the joining of the reinforcing fibers of fiber bundles to each other is cut.

5 **[0034]** The above-described separated bundle aggregate [X] is, for example, as shown in Fig. 8, formed as a separated bundle aggregate [X] having a small width and a predetermined length at a formation of an arbitrary plural formation by being cut at cutting planes 43 oblique with respect to the lengthwise direction of the fiber bundle at a cutting angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) within the separation-processed section 42 of the partially separated fiber bundle 41.

10 **[0035]** As an example of the above-described joined bundle aggregate [Y], the joined bundle aggregate [Y] is, for example, as shown in Fig. 9, formed as a joined bundle aggregate [Y] such as one made with an incision at an end portion in the lengthwise direction of the fiber bundle, by being cut at cutting planes 53 oblique with respect to the lengthwise direction of the fiber bundle at a cutting angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) mainly within the not-separation-processed section 52 of the partially separated fiber bundle [B] 51. Alternatively, the joined bundle aggregate [Y] is, for example, as shown in Fig. 10, formed as a joined bundle aggregate [Y] such as one made with a deep incision at an end portion in the lengthwise direction of the fiber bundle and having an entangled part 63, by being cut at cutting planes 15 65 oblique with respect to the lengthwise direction of the fiber bundle at a cutting angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) over the not-separation-processed section 62 and the separation-processed section 64 having the entangled part 63 at the end portion of the partially separated fiber bundle [B] 61. Alternatively, the joined bundle aggregate [Y] is, for example, as shown in Fig. 11, formed as a joined bundle aggregate [Y] such as one made with a deep incision at an end portion in the lengthwise direction of the fiber bundle and having an entanglement accumulation part 73, by being cut at cutting 20 planes 75 oblique with respect to the lengthwise direction of the fiber bundle at a cutting angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) over the not-separation-processed section 72 and the separation-processed section 74 having the entanglement accumulation part 73 at the end portion of the partially separated fiber bundle [B] 71.

25 **[0036]** Further, the above-described joined cut aggregate [Z] is, for example, as shown in Fig. 12, formed as a joined cut aggregate [Z] small in width and having a relatively large average fiber bundle length in which the end portion in the lengthwise direction is further narrower in width, by being cut at cutting planes 83 oblique with respect to the lengthwise direction of the fiber bundle at a cutting angle  $\theta$  (preferably,  $3^\circ \leq \theta \leq 30^\circ$ ) so as to contain mainly the not-separation-processed section 82 of the partially separated fiber bundle 81 or so as to obliquely traverse the not-separation-processed section 82 over the entire length thereof. In the illustrated example, the not-separation-processed section 82 and the cutting plane 83 at the time of cutting of the partially separated fiber bundle [B] 81 intersect each other, and the joining 30 of single fibers of the fiber bundle 81 to each other is cut off at the intersected part.

35 **[0037]** Where, in the above-described joined cut aggregate [Z], since the average length of the fiber bundle becomes relatively long, at the time of cutting of the fiber bundle or at the time of scattering the aggregates, there is a possibility that cracks are naturally generated even in the not-separation-processed section and aggregates having a smaller number of single fibers may be formed. Such aggregates made into smaller in width are also included in the above-described joined cut aggregate [Z] in the present invention.

40 **[0038]** The chopped fiber bundle [A] comprising the bundled aggregate of discontinuous reinforcing fibers can employ a formation containing at least one kind of aggregate selected from the above-described separated bundle aggregate [X], joined bundle aggregate [Y] and joined cut aggregate [Z]. In the above-described chopped fiber bundle [A], it is preferred that the content of the joined bundle aggregates [Y] is in a range of 0 to 15%, from the viewpoint of exhibiting more excellent mechanical properties and lower variation thereof. Here, the content in the present invention indicates a frequency rate of the joined bundle aggregate [Y] occupied in the chopped fiber bundle [A]. Namely, when the total number of chopped fiber bundles [A] is referred as N(A) and the number of joined bundle aggregates [Y] contained therein is referred as N(Y), it is expressed by the following equation (2).

45 
$$\{N(Y) / N(A)\} \times 100 \cdots (2)$$

50 **[0039]** In the present invention, when producing a fiber-reinforced resin molding material containing the above-described chopped fiber bundle [A], it is desired that the partially separated fiber bundle [B] is cut so as to satisfy the following equation (1) when the above-described chopped fiber bundle [A] is obtained.

$$W \cdot \cos \theta / D \geq 3 \cdots (1)$$

55 W: width of fiber bundle when cutting partially separated fiber bundle  
D: distance between cutting planes in chopped fiber bundle [A]

**[0040]** For example, as shown in Fig. 13, when the cutting angle is referred as  $\theta$ , the width of the fiber bundle at the

time of cutting of the partially separated fiber bundle [B] 91 is referred as  $W$ , and the distance between the cutting planes 92 is referred as  $D$ , the length  $t$  of the side "xy" in  $\Delta xyz$  is

$$t = D / \cos \theta$$

and when the number  $W/t$  for cutting the width  $W$  of the fiber bundle by the cutting planes in the width direction is desired as

$$W/t \geq 3$$

, the aforementioned equation (1) stands from the above-described equation. It is preferred to cut the partially separated fiber bundle so as to satisfy the aforementioned equation (1), because the joined cut aggregate [Z] is effectively made into smaller in width to contribute to improvement of the mechanical properties.

**[0041]** From this equation (1), it is understood that it is effective to increase  $W$  (widen the fiber bundle width) in order to cut the joined bundle aggregate [Y] into small pieces. At this time, because by increasing  $W$ , the thickness of the chopped fiber bundle [A] obtained by cutting is reduced, the flatness of the chopped fiber bundle [A] can be increased, and therefore, because stress concentration at the end portion of the chopped fiber bundle [A] in the molded article is alleviated and the uniformity of the distribution of the chopped fiber bundle [A] and the matrix resin is improved, it is preferred also from the viewpoint of easily exhibiting excellent mechanical properties. However, if the value of  $W$  is too large, there is a possibility that the gathering power of the single fibers forming the fiber bundle decreases, and when cutting the partially separated fiber bundle, the form as a chopped fiber bundle cannot be maintained, breakage of single fibers is liable to occur, and therefore, when made into a fiber-reinforced resin molding material containing the aforementioned random mat and a matrix resin, the flowability may be lowered during molding thereof. Therefore,  $W$  is preferably in a range of  $5 \text{ mm} \leq W \leq 100 \text{ mm}$ , more preferably  $5 \text{ mm} \leq W \leq 50 \text{ mm}$ . Further, it is also good to reduce the cutting angle  $\theta$ . However, there is a limit from the bundle form retention and processability. Further, in order to satisfy the above-described equation (1), although it can be controlled also by the distance  $D$  of the cutting planes, because there is a possibility that the fiber length may fluctuate, it is good to basically set  $D$  as a fixed value so as to be able to cut at a target fiber length.

**[0042]** Thus, in the present invention, by having the chopped fiber bundle [A] comprising the specific bundled aggregate of discontinuous reinforcing fibers, which is formed by cutting the partially separated fiber bundle [B] prepared by alternately forming separation-processed sections and not-separation-processed sections, obliquely with respect to the lengthwise direction of the fiber bundle, while the flowability during molding can be improved, extremely high mechanical properties (strength, elastic modulus) when made into a molded article can be realized, and variations in the mechanical properties can be suppressed to be small.

**[0043]** Next, several embodiments of the chopped fiber bundle [A] used in the present invention, obtained by cutting the above-described partially separated fiber bundle [B], will be explained with reference to the drawings.

**[0044]** Fig. 14 is a plan view of an example of the chopped fiber bundle [A] used in the present invention. In Fig. 14, the chopped fiber bundle [A] CFB1 used in the present invention comprises a large number of reinforcing fibers 111 arranged in one direction and a sizing agent (not shown) for collecting the large number of reinforcing fibers 111 as a bundle. The fiber length  $L_f$  of each reinforcing fiber 111 is, for example, 5 to 100 mm.

**[0045]** The chopped fiber bundle [A] CFB1 has a first transition section 113a in which the number of the reinforcing fibers in a cross section of the fiber bundle in a direction perpendicular to the arrangement direction of the reinforcing fibers 111 (the lengthwise direction of the fiber bundle before cutting, which is the same in the following) is increased from a first tip 112a which is one tip in the arrangement direction of the reinforcing fibers 111 toward a second tip 112b which is the other tip. Further, it has a second transition section 113b in which the number of the reinforcing fibers in the cross section of the fiber bundle is increased from the second tip 112b toward the first tip 112a.

**[0046]** In Fig. 14, the arrangement direction of the reinforcing fibers 111 is depicted in the vertical direction of the figure. The arrangement direction of reinforcing fibers 111 is also the lengthwise direction of chopped fiber bundle [A] CFB1. In Fig. 14, the direction perpendicular to the arrangement direction of the reinforcing fibers 111 is depicted in the left/right direction of the figure. The direction perpendicular to the arrangement direction of the reinforcing fibers 111 is also the width direction of the chopped fiber bundle [A] CFB1.

**[0047]** Between the first transition section 113a and the second transition section 113b, along the arrangement direction of the reinforcing fibers 111, an unchanging section 114 in which the number of reinforcing fibers 111 in the cross section of the fiber bundle is invariable is provided. One end surface 114Ea of the unchanging section 114 coincides with a first terminal end surface 113Ea, which is the terminal end at a side opposite to the first tip 112a of the first transition section 113a. Further, the other end surface 114Eb of the unchanging section 114 coincides with the second terminal end surface 113Eb, which is the terminal end at a side opposite to the second tip 112b of the second transition section 113b.

**[0048]** In the chopped fiber bundle [A] CFB1, the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip 112a and the second tip 112b is set at  $0.05 \text{ mm}^2$  or less per a length of 1 mm in the arrangement direction of the reinforcing fibers 111.

**[0049]** Fig. 14 is a plan view in which a state in which the width in the direction perpendicular to the arrangement direction of the reinforcing fibers 111 of the chopped fiber bundle [A] CFB1 becomes the maximum is depicted. The chopped fiber bundle [A] CFB1 has the maximum width  $W_b$  in the entire area of the unchanging section 114. In the position (section) having the maximum width  $W_b$ , the number of reinforcing fibers 111 is maximized.

**[0050]** In Fig. 14, each of the sides 115a, 115b from the tip to the terminal end in the outer shape of the transition section of both the first transition section 113a and the second transition section 113b is formed with a straight line segment along the arrangement direction of the reinforcing fibers 111, and each of the other sides 116a, 116b is formed with a straight line segment in which the cut ends of a number of reinforcing fibers 111 cut when the chopped fiber bundle [A] CFB1 is manufactured are aligned.

**[0051]** The distance in the width direction of the chopped fiber bundle [A] CFB1 between the side 115a and the side 115b of the chopped fiber bundle [A] CFB1 is the passing width  $W_d$  of the chopped fiber bundle [A] CFB1, and the distance in the lengthwise direction of the chopped fiber bundle [A] CFB1 between the first tip 112a and the second tip 112b is the passing length  $L_d$  of the chopped fiber bundle [A] CFB1.

**[0052]** Fig. 15 is a plan view of another example of the chopped fiber bundle [A] used in the present invention. In Fig. 15, the chopped fiber bundle [A] CFB2 used in the present invention has a similar form to the chopped fiber bundle [A] CFB1 in Fig. 14. Therefore, each part of the chopped fiber bundle [A] CFB2 in Fig. 15 is given with the same symbol as that of the part corresponding to that of the chopped fiber bundle [A] CFB1 in Fig. 14.

**[0053]** The difference between the chopped fiber bundle [A] CFB2 of Fig. 15 and the chopped fiber bundle [A] CFB1 of Fig. 14 is in that the width  $W_b$  of the chopped fiber bundle [A] in the unchanging section 114 of the chopped fiber bundle [A] CFB2 in Fig. 15, that is, the passing width  $W_d$  of the chopped fiber bundle [A] CFB2 is narrower than the passing width  $W_d$  of the chopped fiber bundle [A] CFB1 in Fig. 14. As a result, the length of the side 116a where the cut ends of the multiple reinforcing fibers 111 in the first transition section 113a of the chopped fiber bundle [A] CFB2 in Fig. 15 are arranged is shorter than the length of the side 116a of the chopped fiber bundle [A] CFB1 in Fig. 14, and the length of the side 116b where the cut ends of the multiple reinforcing fibers 111 in the second transition section 113b of the chopped fiber bundle [A] CFB2 in Fig. 15 are arranged is shorter than the length of the side 116b of the chopped fiber bundle [A] CFB1 in Fig. 14.

**[0054]** The fiber length  $L_f$  of each reinforcing fiber 111 in the chopped fiber bundle [A] CFB2 is 5 to 100 mm in this embodiment. In the chopped fiber bundle [A] CFB2, the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip 112a and the second tip 112b is  $0.05 \text{ mm}^2$  or less per a length of 1 mm in the arrangement direction of the reinforcing fibers 111.

**[0055]** Fig. 16 is a plan view showing a further example of the chopped fiber bundle [A] used in the present invention. In Fig. 16, the chopped fiber bundle [A] CFB3 used in the present invention has the same formation as that of the chopped fiber bundle [A] CFB1 in Fig. 14 except a point in that it does not have the unchanging section 114 of the chopped fiber bundle [A] CFB1 in Fig. 14. Therefore, each part of the chopped fiber bundle [A] CFB3 in Fig. 16 is given with the same symbol as that of the part corresponding to that of the chopped fiber bundle [A] CFB1 in Fig. 14.

**[0056]** The chopped fiber bundle [A] CFB3 in Fig. 16 comprises a first transition section 113a in which the number of reinforcing fibers 111 increases from the first tip 112a toward the second tip 112b and a second transition section 113b in which the number of reinforcing fibers 111 increases from the second tip 112b toward the first tip 112a. In the chopped fiber bundle [A] CFB3, the first terminal end surface 113Ea, which is the terminal end at a side opposite to the first tip 112a of the first transition section 113a, and the second terminal end surface 113Eb, which is the terminal end at a side opposite to the second tip 112b of the second transition section 113b, directly coincide with each other.

**[0057]** The chopped fiber bundle [A] CFB3 has a maximum width  $W_b$  at a portion where the both terminal end surfaces 113Ea and 113Eb coincide with each other. In the position (section) having the maximum width  $W_b$ , the number of reinforcing fibers 111 is maximized. Further, since these both terminal end surfaces 113Ea and 113Eb coincide with each other, the value of the passing length  $L_d$  of the chopped fiber bundle [A] CFB3 is twice the value of the length  $L_f$  of the reinforcing fiber 111.

**[0058]** The fiber length  $L_f$  of each reinforcing fiber 111 in the chopped fiber bundle [A] CFB3 is 5 to 100 mm in this embodiment. In the chopped fiber bundle [A] CFB3, the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip 112a and the second tip 112b is set to  $0.05 \text{ mm}^2$  or less per 1 mm in the arrangement direction of the reinforcing fibers 111.

**[0059]** Although as the cross-sectional shape of the chopped fiber bundle [A] in the direction perpendicular to the lengthwise direction, various shapes such as a circle, an ellipse or a square can be employed, from the viewpoints of stability of the cross-sectional shape of the chopped fiber bundle, good handling of the chopped fiber bundle, and ease of manufacture of the chopped fiber bundle, the cross-sectional shape of the chopped fiber bundle is preferably a circle, an ellipse or a square, and particularly preferably it is a flat rectangle or a flat ellipse.

**[0060]** Fig. 17 shows a list of the respective plan views ((a) to (g)) of other seven examples of the chopped fiber bundle [A] used in the present invention. Each chopped fiber bundle [A] in Fig. 17 comprises a number of reinforcing fibers 111 aligned in the vertical direction in the figure and collected as a bundle by a sizing agent.

**[0061]** The chopped fiber bundle [A] CFB5a in Fig. 17(a) has four tips on the upper side and four tips on the lower side, and has a V-shaped notch between adjacent tips. The outer shape of the chopped fiber bundle [A] CFB5a consists of 16 sides, and all the sides consist of straight line segments.

**[0062]** The chopped fiber bundle [A] CFB5b in Fig. 17(b) has one tip on the upper side and two tips on the lower side, and has a V-shaped notch between the lower-side two tips. The outer shape of the chopped fiber bundle [A] CFB5b comprises six sides, and each side consists of a straight line segment.

**[0063]** The chopped fiber bundle [A] CFB5c in Fig. 17(c) has one tip on the upper side and one tip on the lower side. The outer shape of the chopped fiber bundle [A] CFB5c comprises four sides, two of which are composed of curved line segments and the other two sides are composed of straight line segments.

**[0064]** The chopped fiber bundle [A] CFB5d in Fig. 17(d) has two tips on the upper side and one tip on the lower side. The outer shape of the chopped fiber bundle [A] CFB5d comprises four sides, of which the side connecting the upper-side two tips consists of a U-shaped curved line segment, the side including the lower-side tip is consists of a U-shaped curved line segment, and the remaining two sides consist of straight line segments.

**[0065]** The chopped fiber bundle [A] CFB5e in Fig. 17(e) has one tip on the upper side and one tip on the lower side. The outer shape of the chopped fiber bundle [A] CFB5e comprises two sides, and their sides are composed of outwardly convex curved line segments connecting the upper tip and the lower tip, respectively.

**[0066]** The chopped fiber bundle [A] CFB5f in Fig. 17(f) has one tip on the upper side and one tip on the lower side. The outer shape of the chopped fiber bundle [A] CFB5f comprises six sides, and each side consists of a straight line segment.

**[0067]** The chopped fiber bundle [A] CFB5g in Fig. 17(g) has one tip on the upper side and one tip on the lower side. The outer shape of the chopped fiber bundle CFB5g comprises four sides, and each side consists of a straight line segment..

**[0068]** The chopped fiber bundle [A] used in the present invention is used for manufacturing a molding material for molding a fiber reinforced molded body (fiber reinforced plastic). This molding material comprises aggregates of a large number of the aforementioned chopped fiber bundles [A]. In case where a molded body having a complicated shape is formed by using this molding material, a good molding followability to the complicated shape is required. When the fiber length  $L_f$  of all the reinforcing fibers 111 contained in the chopped fiber bundles [A] used in the present invention is set to 100 mm or less, the molding material comprising a number of the chopped fiber bundles [A] used in the present invention has a good molding followability.

**[0069]** In case where the fiber length  $L_f$  exceeds 100mm, as the fiber length becomes longer, the reinforcing fibers 111 hardly flow in the arrangement direction thereof in the molding process of a molded body, and it becomes difficult to produce a molded body having a complicated shape. In case where the fiber length  $L_f$  is less than 5 mm, although the flowability of the reinforcing fibers 111 in the molding process of a molded body is improved, the mechanical properties of the molded body to be obtained decrease. More preferably, the length  $L_f$  of each reinforcing fiber 111 in the chopped fiber bundle [A] used in the present invention is 10 to 50 mm from the relationship between the flowability of the reinforcing fibers in the molding process of the molded body and the mechanical properties of the obtained molded body.

**[0070]** The smaller the number of reinforcing fibers with a fiber length less than 5mm contained in the chopped fiber bundle [A] is, the better it is, and it is preferably less than 5% of the total number of reinforcing fibers forming the chopped fiber bundle. Namely, in the present invention, the condition where the fiber length  $L_f$  of reinforcing fibers 111 forming a chopped fiber bundle is 5 to 100mm includes a condition where the number of reinforcing fibers having a fiber length less than 5 mm is 5% or less of the total number of reinforcing fibers forming the chopped fiber bundle and the fiber length of all reinforcing fibers is 100mm or less.

**[0071]** Generally, a fiber reinforced plastic (hereinafter, also referred to as "short fiber reinforced plastic") is produced by molding a molding material comprising aggregates of a large number of chopped fiber bundles. In case where a load is applied to the fiber reinforced plastic, most of the load is to be received by the reinforcing fibers present in the fiber reinforced plastic. In case of a chopped fiber bundle, the multiple reinforcing fibers forming it are in a state of being cut with a certain length. Therefore, it needs that the load having been received by the reinforcing fibers of a certain chopped fiber bundle is delivered from the end portion of the chopped fiber bundle via a matrix resin to the reinforcing fibers of another chopped fiber bundle located nearby at the end portion of the chopped fiber bundle.

**[0072]** In the chopped fiber bundle [A] used in the present invention, by making the number of reinforcing fibers forming the chopped fiber bundle [A] smaller at both end portions of the chopped fiber bundle [A] than at the center thereof, the load assigned to the chopped fiber bundle [A], which is the largest at the central portion of the chopped fiber bundle [A], is gradually delivered to chopped fiber bundles positioned nearby, little by little, towards the end portions of the chopped fiber bundle [A] through the reinforcing fibers whose number is decreasing, and therefore, in the fiber reinforced plastic comprising the chopped fiber bundles [A] used in the present invention, stress concentration is unlikely to occur.

**[0073]** Therefore, in case of the chopped fiber bundle [A] used in the present invention, as compared with the conventional case of the chopped fiber bundle in which all the reinforcing fibers are cut at the same position, the strength of the fiber reinforced plastic to be obtained is remarkably improved. Not only that, because stress concentration does not occur, initial damage (crack) is unlikely to occur. In fiber reinforced plastics applications, there are some applications that cannot be applied because sounds are caused by initial damage and induce anxiety, but even in such applications, the fiber reinforced plastic comprising the chopped fiber bundle [A] used in the present invention (the fiber reinforced plastic of the present invention) becomes possible to be used. Further, although initial damage largely affects fatigue strength, in case of the fiber reinforced plastic of the present invention, because initial damage is small, not only static strength but also fatigue strength is greatly improved. In addition, since the chopped fiber bundle [A] used in the present invention is obtained by cutting the partially separated fiber bundle [B], the stress concentration is further made less likely to occur.

**[0074]** With respect to the increase in the number of reinforcing fibers 111 in the transition sections 113a and 113b of the chopped fiber bundle [A] used in the present invention, in case where there are increases in the number of reinforcing fibers 111 at least at two places in the transition sections 113a and 113b and the maximum value of the total cross-sectional area of reinforcing fibers in the cross section of the chopped fiber bundle [A] at the number increasing places is  $0.008 \text{ mm}^2$  or less, the increase in the number of reinforcing fibers 111 in the transition sections 113a and 113b can be said to be a continuous increase. It is preferred that the total cross-sectional area of the reinforcing fibers in the cross section of the chopped fiber bundle [A] in the above-described number increasing places is  $0.002 \text{ mm}^2$  or less, from the viewpoint that the more smoothly the number of reinforcing fibers is increased, the more unlikely the above-described stress concentration is to occur.

**[0075]** Over the entire range in the lengthwise direction of the chopped fiber bundle [A] used in the present invention including transition sections 113a and 113b in which the number of reinforcing fibers 111 actually changes (the entire range of the passing length  $L_d$ ), the change amount of the total cross-sectional area of reinforcing fibers is set at  $0.05 \text{ mm}^2$  or less per a length of 1 mm. By the provision of this change amount, the above-described stress concentration can be effectively prevented. This change amount is preferably  $0.04 \text{ mm}^2$  or less per a length of 1 mm, more preferably  $0.025 \text{ mm}^2$  or less.

**[0076]** The total cross-sectional area of reinforcing fibers at an arbitrary position in the chopped fiber bundle [A] is the total sum obtained by adding the cross-sectional areas of respective reinforcing fibers of all reinforcing fibers existing in a plane (cross section) orthogonal to the arrangement direction of the reinforcing fibers.

**[0077]** Fig. 18 shows a plan view (a) of the chopped fiber bundle [A] CFB2 used in the present invention of Fig. 15, a side view (b), and a graph (c) showing the state of increase/decrease in number of reinforcing fibers 111 in the arrangement direction of the reinforcing fibers 111 of the chopped fiber bundle [A] CFB2. In the graph of Fig. 18(c), the horizontal axis X represents the position in the passing length  $L_d$  of the chopped fiber bundle [A] CFB2, and the vertical axis Y represents the number of the reinforcing fibers 111 or the total cross-sectional area of the reinforcing fibers 111.

**[0078]** As shown in the graph of Fig. 18(c), the number of reinforcing fibers 111 of the chopped fiber bundle [A] CFB2 continuously increases from the first tip 112a toward the first terminal end surface 113Ea of the first transition section along the lengthwise direction of the chopped fiber bundle [A] CFB2, and becomes a constant value at the first terminal end surface 113Ea. This constant value is maintained in the unchanging section 114 from the first terminal end surface 113Ea to the second terminal end surface 113Eb of the second transition section. The number of reinforcing fibers 111 then decreases continuously from the second terminal end surface 113Eb toward the second tip 112b along the lengthwise direction of the chopped fiber bundle [A] CFB2. The number of reinforcing fibers 111 in the unchanging section 114 is the maximum value of the number of reinforcing fibers 111 in the chopped fiber bundle [A] CFB2.

**[0079]** Fig. 19 shows a plan view (a) of the chopped fiber bundle [A] CFB3 used in the present invention of Fig. 16, a side view (b), and a graph (c) showing the state of increase/decrease in number of reinforcing fibers 111 in the arrangement direction of the reinforcing fibers 111 of the chopped fiber bundle [A] CFB3. In the graph of Fig. 19(c), the horizontal axis X represents the position in the passing length  $L_d$  of the chopped fiber bundle [A] CFB3, and the vertical axis Y represents the number of the reinforcing fibers 111 or the total cross-sectional area of the reinforcing fibers 111.

**[0080]** As shown in the graph of Fig. 19(c), the number of reinforcing fibers 111 of the chopped fiber bundle [A] CFB3 continuously increases from the first tip 112a toward the first terminal end surface 113Ea of the first transition section along the lengthwise direction of the chopped fiber bundle [A] CFB3. The chopped fiber bundle [A] CFB3 does not have an unchanging section in which the number of reinforcing fibers is maintained at a constant value in the lengthwise direction of the fiber bundle, and the first terminal end surface 113Ea and the second terminal end surface 113Eb of the second transition section 113b coincide with each other, and therefore, the number of reinforcing fibers 111 in the first terminal end surface 113Ea (second terminal end surface 113Eb) indicates the maximum value. The number of reinforcing fibers 111 then decreases continuously from the second terminal end surface 113Eb toward the second tip 112b along the lengthwise direction of the chopped fiber bundle [A] CFB3.

**[0081]** The change in the number of reinforcing fibers in the lengthwise direction of the chopped fiber bundle [A] used in the present invention has two formations the first formation in which it becomes a constant value after the increase,

and decreases thereafter, and the second formation in which after the increase, it decreases without having a constant value.

**[0082]** In the chopped fiber bundle [A] used in the present invention, the change amount CFTS of the total cross-sectional area FTS of the reinforcing fiber 111 in the cross section of the chopped fiber bundle [A] between the first tip 112a and the second tip 112b (exemplified in Figs. 18 and 19) is  $0.05 \text{ mm}^2$  or less per a length of 1 mm in the arrangement direction of the reinforcing fibers 111. The total cross-sectional area FTS of the reinforcing fibers 111 in the cross section of the chopped fiber bundle [A] is the total sum of the cross sectional areas of the respective reinforcing fibers 111 existing in the cross section.

**[0083]** In case where the cross-sectional areas of respective reinforcing fibers 111 present in the cross section of the chopped fiber bundle [A] vary in a range of  $\pm 10\%$  or less relative to the cross-sectional area of the representative reinforcing fiber selected among them, as the total cross-sectional area FTS of the reinforcing fibers 111, a value obtained by multiplying the number of reinforcing fibers 111 present in the cross section by the cross-sectional area of the representative reinforcing fiber is used. Further, in case where the maximum width  $W_b$  of the chopped fiber bundle is less than 3 mm, as the change amount CFTS of the total cross-sectional area FTS of the reinforcing fiber, a value obtained by dividing the maximum value of the total cross-sectional area FTS of the reinforcing fibers 111 in the chopped fiber bundle [A] by the length (mm) of the transition sections 113a and 113b in the arrangement direction of the reinforcing fibers is used.

**[0084]** Figs. 14 to 17 show various examples of the chopped fiber bundle [A] used in the present invention. Any of these chopped fiber bundles [A] has a transition section in which the number of reinforcing fibers increases and the change amount of total cross-sectional area of reinforcing fibers is  $0.05 \text{ mm}^2$  or less per 1 mm in the arrangement direction of the reinforcing fibers over the entire range of the lengthwise direction of the chopped fiber bundle. In the chopped fiber bundle [A] used in the present invention, the maximum value of the total cross-sectional area of reinforcing fibers is preferably  $0.1 \text{ mm}^2$  or more.

**[0085]** The state in which the number of reinforcing fibers increases from the tip to the terminal end of the transition section of the chopped fiber bundle [A] used in the present invention can be expressed conversely as a state in which the number of reinforcing fibers decreases from the central portion of the chopped fiber bundle toward the tip of the chopped fiber bundle. By this decreasing state of the number of reinforcing fibers, the above-described occurrence of stress concentration in the fiber reinforced plastic is prevented. This decreasing state of the number of reinforcing fibers is preferably a formation in which the number of reinforcing fibers gradually decreases, that is, continuously decreases. If the chopped fiber bundle is thicker, the number of reinforcing fibers is larger, and the total cross-sectional area of reinforcing fibers is larger, the effect of preventing the occurrence of stress concentration is more improved. The larger the total cross-sectional area of the reinforcing fibers is, the larger the load born by one chopped fiber bundle in the fiber reinforced plastic becomes, but even if the load to be born is large, the state of being delivered with the load at a stroke to the end portion of an adjacent chopped fiber bundle through a matrix resin can be prevented by the state decreased with the number of reinforcing fibers in the transition section. Namely, the transmission of the load between adjacent chopped fiber bundles [A] is gradually performed by the decreasing state of the number of reinforcing fibers in the transition section, and the stress concentration at the end portion of the chopped fiber bundle [A] can be prevented.

**[0086]** In case where a chopped fiber bundle [A] is produced, production of a thicker chopped fiber bundle is more excellent in processability and the production cost also becomes lower. However, in case where a conventional chopped fiber bundle cut in a direction orthogonal to the lengthwise direction of a fiber bundle is made as a thick chopped fiber bundle, the conventional short fiber reinforced plastic molded using this thick chopped fiber bundle is low in strength. Therefore, this short fiber reinforced plastic has a problem that it is difficult to be applied to a member for strength.

**[0087]** Even if the chopped fiber bundle [A] used in the present invention is a thick chopped fiber bundle, the short fiber reinforced plastic formed by using it has a high strength as compared with a conventional short fiber reinforced plastic formed by using a conventional thick chopped fiber bundle. Therefore, it is possible to lower the production cost of the chopped fiber bundle, and it also becomes possible to produce a short fiber reinforced plastic having a high strength. From the viewpoint of a thick chopped fiber bundle, the maximum value of the total cross-sectional area of the reinforcing fibers is preferably  $0.2 \text{ mm}^2$  or more. Where, from the viewpoint of the degree of freedom of thickness design when made into a fiber reinforced plastic, the maximum value of the total cross-sectional area of the reinforcing fibers is preferably  $30 \text{ mm}^2$  or less, more preferably  $5 \text{ mm}^2$  or less.

**[0088]** On the other hand, in case where the maximum value of the total cross-sectional area of reinforcing fibers is less than  $0.1 \text{ mm}^2$ , it is preferred that the change amount of the number of reinforcing fibers is 30% or less of the maximum number of reinforcing fibers (the maximum value of the number) per 1 mm in the arrangement direction of the reinforcing fibers over the entire range of the chopped fiber bundle [A]. In where the maximum value of the total cross-sectional area of reinforcing fibers is less than  $0.1 \text{ mm}^2$ , namely, even if it is a thin chopped fiber bundle [A], as compared with the formation that a load is released at a stroke as in the conventional chopped fiber bundle, in the formation that the number of reinforcing fibers gradually decreases from the central portion of the chopped fiber bundle toward the tip is preferable because the transmission of a load in the fiber reinforced plastic is gradually performed.

**[0089]** In the chopped fiber bundle [A] used in the present invention, it is also preferred that the fiber lengths  $L_f$  of respective reinforcing fibers 111 are the same. When producing the chopped fiber bundle [A], if the fiber lengths of respective reinforcing fibers are the same, since it is possible to produce chopped fiber bundles by cutting a continuous reinforcing fiber bundle at a same interval in the lengthwise direction, the production efficiency of the chopped fiber bundles is good, and when integrating a number of chopped fiber bundles [A] to make a molding material and performing molding of a molded body using this molding material, if the fiber lengths of respective reinforcing fibers are the same, the control of the flow of the reinforcing fibers is easier. Examples of chopped fiber bundles used in the present invention where the fiber lengths of respective reinforcing fibers are the same are shown in Figs. 14, 15, 16, 17(a), 17(b), 17(c) and 17(d).

**[0090]** The condition where the fiber lengths of respective reinforcing fibers are the same means that reinforcing fibers having a fiber length within the range of  $\pm 5\%$  from the average value of the fiber lengths of the reinforcing fibers contained in the chopped fiber bundle [A] occupy 95% of the total reinforcing fibers contained in the chopped fiber bundle.

**[0091]** As the reinforcing fiber used for the chopped fiber bundle [A] used in the present invention, the fiber kind is not particularly limited as long as it is a fiber bundle comprising a plurality of reinforcing fibers. For example, there are an organic fiber such as aramid fiber, polyethylene fiber, and polyparaphenylene benzoxadole

**[0092]** (PBO) fiber, an inorganic fiber such as glass fiber, carbon fiber, silicon carbide fiber, alumina fiber, tyranno fiber, basalt fiber and ceramic fiber, a metal fiber such as stainless steel fiber and steel fiber, and as others, boron fiber, natural fiber, and modified natural fiber. Further, it may be a combination of two or more reinforcing fibers of different kinds. Among these fibers, since carbon fiber is lightweight, has excellent specific strength and specific elastic modulus, and further has excellent heat resistance and chemical resistance, it is preferably used as the reinforcing fiber. A molded body (fiber reinforced plastic) produced from chopped fiber bundles comprising carbon fibers used in the present invention is suitably used for a member such as an automobile panel desired with reduction in weight.

**[0093]** In the chopped fiber bundle [A] used in the present invention, it is preferred that the reinforcing fiber is a carbon fiber, the number of carbon fibers is 1,000 to 700,000, and over the entire range of the chopped fiber bundle, and the change amount of the number of the carbon fibers is 1,400 or less each time it moves by 1 mm in the arrangement direction of the carbon fibers.

**[0094]** The carbon fiber is preferably a polyacrylonitrile-based carbon fiber which is easy to obtain a high strength. Considering that the diameter of single fiber of easily obtainable carbon fiber is about 5 to 10  $\mu\text{m}$ , it is preferred that the number of carbon fibers in the chopped fiber bundle is 1,000 to 700,000. The number of the carbon fibers is more preferably 1,000 to 100,000. Since a continuous carbon fiber bundle having a high strength and having 6,000 to 50,000 fibers is inexpensive and easy to obtain, it is preferably used when producing the chopped fiber bundle [A] used in the present invention. The continuous carbon fiber bundle as described above is supplied as a yarn wound body (package) wound with the fiber bundle on a bobbin. Although the fiber bundle is preferred to be untwisted, a strand with twists can also be used, and even if twisting occurs during conveyance, it can be applied to the present invention. Further, in case of using so-called large tow with a large number of single fibers bundled, since the price per unit weight of fiber bundle is inexpensive, as the number of single fibers is larger, the cost of the final product can be reduced preferably. Further, as a large tow, a so-called doubling form in which fiber bundles are wound together in a form of one bundle may be used.

**[0095]** When the reinforcing fiber as described above is used, it is preferred that the reinforcing fiber is subjected to a surface treatment for the purpose of improving the adhesiveness with a matrix resin [M] or the like. As the method for the surface treatment, there are electrolytic treatment, ozone treatment, ultraviolet treatment and the like.

**[0096]** The fiber bundle used in the present invention is preferably in a state of being bundled in advance. Here, the state being bundled in advance means, for example, a state bundled due to entanglement of reinforcing fibers constituting the fiber bundle with each other, a state bundled by a sizing agent applied to the fiber bundle, and a state bundled due to twists being contained in the production process of the fiber bundle.

**[0097]** If the change amount of the number of reinforcing fibers is not more than 1,400 every time 1 mm movement in the arrangement direction of reinforcing fibers over the entire range of chopped fiber bundle [A] used in the present invention, stress concentration can be effectively prevented in a fiber reinforced plastic. The change amount of the number of reinforcing fibers is preferably 1,000 or less. In order to improve the strength of the fiber reinforced plastic, the change amount of the number of reinforcing fibers is preferably 600 or less, more preferably 300 or less.

**[0098]** In case where the maximum width  $W_b$  of chopped fiber bundle [A] is less than 3 mm, as the change amount of the number of reinforcing fibers, a value calculated by dividing the maximum value of the number of reinforcing fibers of the chopped fiber bundle [A] by the length of the transition section in the arrangement direction of the reinforcing fibers and proportionally converting it to a change amount per 1 mm is used. In this case, there are increases in the number of reinforcing fibers in at least two places within the transition section, and the number of reinforcing fibers included in the cross section of the chopped fiber bundle [A] at the places where the number of reinforcing fibers increases is preferably 200 or less, and more preferably 50 or less.

**[0099]** In the chopped fiber bundle [A] used in the present invention, the ratio  $W_b/T_b$  of the maximum width  $W_b$  to the maximum thickness  $T_b$  is preferably 20 to 400. The ratio  $W_b/T_b$  represents the flatness ratio of the chopped fiber bundle.

The larger the flatness ratio is, the flatter the chopped fiber bundle is. A flat chopped fiber bundle [A] brings about an increase in the strength of a fiber reinforced plastic. In particular, as the chopped fiber bundle [A] is thicker, that is, the number of single fibers constituting the chopped fiber bundle is larger, the contribution of the flatness ratio to the strength improving effect becomes larger. The value of the maximum thickness  $T_b$  is preferably 150  $\mu\text{m}$  or less, and more preferably 100  $\mu\text{m}$  or less.

**[0100]** A flat chopped fiber bundle [A] can be produced, for example, by opening a continuous reinforcing fiber bundle (for example, a partially separated fiber bundle [B]) drawn out in one direction and then cutting it. This opening of the continuous reinforcing fiber bundle can be carried out, for example, by passing the continuous reinforcing fiber bundle in contact with a roller, vibrating the continuous reinforcing fiber bundle, or performing air blowing to the continuous reinforcing fiber bundle. Here, in case of performing the opening treatment to the partially separated fiber bundle [B], the opening treatment may be performed by subjecting the fiber bundle to a separation processing, once winding it up as a partially separated fiber bundle [B] and then performing the opening treatment, and alternatively, may be performed in the process for obtaining the partially separated fiber bundle [B]. Particularly, in the process for obtaining the partially separated fiber bundle [B], by performing the opening treatment to the continuous reinforcing fiber bundle and performing a separation processing at a state performed with opening treatment, more uniform separation can be carried out, and further, from the viewpoint that it is possible to reduce the risk of fluffs and yarn breakage occurring at the time of separation processing, it is more preferred to perform the opening treatment in the process for obtaining the partially separated fiber bundle [B]. Further, from the viewpoint of avoiding narrowing of the width of the fiber bundle once opened, it is further preferred to perform the opening treatment in the process for obtaining the partially separated fiber bundle [B], and to subject it to a cutting step without winding it, to obtain a chopped fiber bundle [A].

**[0101]** A particularly preferred formation of the chopped fiber bundle [A] used in the present invention is a formation in which the end portion of the chopped fiber bundle has a side that extends obliquely with respect to the arrangement direction of the reinforcing fibers. A formation is more preferred wherein the oblique side is linearly formed at an angle of 3 to 30° with respect to the arrangement direction of the reinforcing fibers.

**[0102]** The chopped fiber bundle [A] used in the present invention having such a formation can be manufactured, for example, by drawing a continuous reinforcing fiber bundle (the partially separated fiber bundle [B]) in one direction, and cutting the continuous reinforcing fiber bundle drawn out linearly at an angle of 3 to 30° with respect to the arrangement direction of the reinforcing fibers (the drawing direction of the continuous reinforcing fiber bundle) so that the fiber length of the reinforcing fibers becomes 5 to 100 mm. In this manufacturing method, a flatter chopped fiber bundle [A] can be produced by cutting the opening the continuous reinforcing fiber bundle drawn out in one direction after opening it. Although the conventional chopped fiber bundle has been produced by cutting a continuous reinforcing fiber bundle in the direction perpendicular to the arrangement direction of the reinforcing fibers (the drawing direction of the continuous reinforcing fiber bundle), only by cutting the continuous reinforcing fiber bundle at an angle of 3 to 30° with respect to the arrangement direction of the reinforcing fibers (the drawing direction of the continuous reinforcing fiber bundle), the chopped fiber bundle [A] used in the present invention which enables production of a fiber reinforced plastic having a high strength can be obtained.

**[0103]** The smaller the angle of the side formed by the cut reinforcing fiber arrangement at the end portion of the chopped fiber bundle [A] with respect to the arrangement direction of the reinforcing fibers is, the higher the increasing effect of the strength of the fiber reinforced plastic molded by using it becomes. In case where the angle is 30° or less, its effect is remarkable. On the other hand, however, the handling ability of the chopped fiber bundle itself is reduced. Further, the smaller the angle between the arrangement direction of the reinforcing fibers and a cutting blade is, the lower the stability in the cutting process becomes. Therefore, the angle is preferably 3° or more. More preferably, the angle is 4 to 25°. It is further preferred that the angle is 5 to 15° in consideration of a good balance between achievement of a high strength of a fiber reinforced plastic and the processability in the manufacturing process of the chopped fiber bundle. Where, the angle mentioned here is represented by an absolute value.

**[0104]** The chopped fiber bundles [A] used in the present invention shown in Figs. 14, 15 and 16 are manufactured by cutting a continuous reinforcing fiber bundle (partially separated fiber bundle [B]) in the lengthwise direction thereof at an identical cutting interval. The chopped fiber bundle [A] CFB1 used in the present invention in Fig. 14 is obtained by cutting a relatively wide continuous reinforcing fiber bundle, and has a formation in which the lengths of the sides 116a and 116b on which the cut ends of the reinforcing fibers 111 are arranged are long. Since the lengths of the sides 116a and 116b are long, the reinforcing fibers are easy to be opened at the time of manufacturing a molding material or molding a molded body using the molding material. Therefore, the thickness of each chopped fiber bundle [A] in the molding material or the molded body is reduced, and the strength of the obtained molded body (fiber reinforced plastic) is easily improved.

**[0105]** The chopped fiber bundle [A] CFB2 used in the present invention in Fig. 15 is obtained by cutting a continuous reinforcing fiber bundle (partially separated fiber bundle [B]) having a relatively small width, and has a formation in which the lengths of the sides 116a and 116b on which the cut ends of the reinforcing fibers 111 are arranged are short. Since the lengths of the side 116a and 116b are short, the reinforcing fibers are difficult to be spread and excellent in handling

of the chopped fiber bundle [A].

[0106] The chopped fiber bundle [A] CFB3 used in the present invention shown in Fig. 16 does not have an unchanging section existing in the chopped fiber bundle [A] of Fig. 14 or Fig. 15, from the relationship between the cutting angle and the width of the continuous reinforcing fiber bundle when the continuous reinforcing fiber bundle (partially separated fiber bundle [B]) is cut, and comprises substantially only two transition sections 113a and 113b. In this chopped fiber bundle [A] CFB3, the passing length  $L_d$  of the chopped fiber bundle [A] CFB3 is twice the fiber length  $L_f$  of the reinforcing fibers 111.

[0107] As a cutting means for the continuous reinforcing fiber bundle for manufacturing the chopped fiber bundle [A], for example, there is a guillotine cutter or a rotary cutter such as a roving cutter or the like. The continuous reinforcing fiber bundle is inserted into the cutting means and cut in a state in which the lengthwise direction of the continuous reinforcing fiber bundle and the direction of the cutting blade mounted on the cutting means are extended relatively obliquely.

[0108] For the manufacture of the chopped fiber bundle [A] CFB5a in Fig. 17(a), for a notched blade is used, for the manufacture of the chopped fiber bundle [A] CFB5b in Fig. 17(b), a V-shaped blade is used, for the manufacture of the chopped fiber bundle [A] CFB5c in Fig. 17(c), a streamlined blade is used, and for the manufacture of the chopped fiber bundle [A] CFB5d in Fig. 17(d), a U-shaped blade is used, respectively. The chopped fiber bundle [A] CFB5e in Fig. 17(e) is manufactured by cutting a continuous reinforcing fiber bundle obliquely with respect to the lengthwise direction of the continuous reinforcing fiber bundle while applying a sizing agent such as water to the continuous reinforcing fiber bundle, and converging both side portions in the lengthwise direction of the cut piece obtained by the cutting toward both tip portions at a canoe-like form. For manufacture of the chopped fiber bundle [A] CFB5 f in Fig. 17(f) and the chopped fiber bundle [A] CFB5g in Fig. 17(g), a plurality of blades having different shapes are used.

[0109] The chopped fiber bundle [A] used in the present invention can also be manufactured by applying shearing to a chopped fiber bundle, which is obtained by the conventional chopped fiber bundle manufacturing method and in which the cut ends of the reinforcing fibers are arranged in a direction perpendicular to the lengthwise direction of the chopped fiber bundle, in the thickness direction of the chopped fiber bundle, and forming a transition section in which the number of reinforcing fibers changes. Further, the chopped fiber bundle [A] used in the present invention can also be manufactured by spinning a continuous reinforcing fiber bundle using a pull-cut spinning means. The chopped fiber bundle [A] obtained by pull-cutting has a formation in which reinforcing fibers differing in length in the lengthwise direction of chopped fiber bundle [A] are arranged at both ends of the chopped fiber bundle [A], and by the portions, transition sections are formed.

[0110] In the chopped fiber bundle [A] used in the present invention, it is preferred that a number of reinforcing fibers forming it contain a sizing agent for maintaining the state of the bundle. As the sizing agent, one may be employed which can maintain a number of reinforcing fibers as a state of bundle, and the material of which has no problem in compatibility with a resin used for producing a molded body (fiber reinforced plastic) comprising the chopped fiber bundles [A].

[0111] The chopped fiber bundle [A] used in the present invention is manufactured by cutting a continuous reinforcing fiber bundle (partially separated fiber bundle [B]). To this continuous reinforcing fiber bundle, usually, a sizing agent is applied at the stage of manufacturing the continuous reinforcing fiber bundle in order to improve handling ability of the fiber bundle. Therefore, this sizing agent can be used as it is as a bundling agent for the chopped fiber bundle [A] used in the present invention, and in this case, there is an advantage that there is no need to separately prepare another bundling agent.

[0112] When the sizing agent is used as the bundling agent, the amount of the sizing agent applied to the reinforcing fibers is preferably 0.1 to 10% by mass, relative to a standard of the mass of the entire chopped fiber bundle [A]. In case where the same amount of sizing agent as this amount is applied to the continuous reinforcing fiber bundle used for the manufacture of the chopped fiber bundle [A], when cutting the continuous reinforcing fiber bundle, the reinforcing fibers will not break apart from each other, and the shape of the chopped fiber bundle [A] obtained by cutting becomes an intended shape. In case where the chopped fiber bundle [A] is manufactured from the continuous reinforcing fiber bundle, by a condition where 0.1 to 10 mass% of sizing agent is applied to the continuous reinforcing fibers to be used, the processability in the manufacturing process of the chopped fiber bundle [A] is improved drastically. In addition, the handling ability of the chopped fiber bundle [A] when producing a molding material using the chopped fiber bundle [A] is also improved.

[0113] For example, 0.1 to 10 mass% of a sizing agent dissolved or dispersed in a solvent is added to the drawn continuous reinforcing fiber bundle, and after the continuous reinforcing fiber bundle is cut, it is heated to dry the solvent, or after the continuous reinforcing fiber bundle is heated to dry the solvent, it is cut, thereby obtaining the chopped fiber bundle used in the present invention.

[0114] As the sizing agent, for example, there are an epoxy resin, a phenol resin, an unsaturated polyester resin, a vinyl ester resin, a polyamide resin, a urethane resin, or a mixed resin obtained by mixing them. These resins are diluted with water, a solvent or the like, and applied to a continuous reinforcing fiber bundle.

[0115] A fiber-reinforced resin molding material containing a random mat containing the above-described chopped fiber bundles [A] in the present invention and a matrix resin [M] contains a large number of aggregates of the above-

described chopped fiber bundles [A], and Fig. 20 is a plan view showing an example thereof. In Fig. 20, a fiber-reinforced resin molding material 191 according to the present invention contains a large number of aggregates of the aforementioned chopped fiber bundles CFB (for example, chopped fiber bundle [A] CFB3 shown in Fig. 16). Although the fiber-reinforced resin molding material of the present invention contains a large number of aggregates of the aforementioned chopped fiber bundles [A], this aggregate may be combined with another base material, for example, a base material composed of a large number of continuous fibers. In any formation, because the fiber-reinforced resin molding material of the present invention is used for the production of a fiber reinforced plastic, it is preferred that it has a good handling property and a property suitable for pressurization molding such as press molding and drape molding.

**[0116]** The fiber-reinforced resin molding material of the present invention can be produced, for example, by sandwiching a random mat, obtained by spraying a large number of the aforementioned chopped fiber bundles [A] in a sheet form, with matrix resin sheets from upper and lower sides, to form as a molding material in which a large number of the chopped fiber bundles [A] and a matrix resin are integrated. Such a molding material is usually called as an SMC (Sheet Molding Compound) sheet or a stampable sheet.

**[0117]** The amount of the matrix resin [M] in the fiber-reinforced resin molding material of the present invention is preferably 20 to 75% by mass. In case where the amount of the matrix resin in the molding material is less than 20% by mass, because the resin amount is small, there is a case where the flowability, which is one of the major characteristics of the molding material of the present invention, may be impaired. In case where the amount of the matrix resin in the molding material is more than 75% by mass, because the amount of reinforcing fibers is smaller than the amount of the resin, it becomes difficult to improve the mechanical properties of the resulting fiber-reinforced plastic. The amount of the matrix resin in the molding material is more preferably 35 to 55% by mass.

**[0118]** As the matrix resin [M] used for the fiber-reinforced resin molding material, there is a case where a thermosetting resin is preferable. Since a thermosetting resin has a crosslinked structure, in general, it has a high elastic modulus and an excellent shape stability. In a fiber-reinforced plastic produced by this, a high elastic modulus and a good dimensional stability are exhibited. In the thermosetting resin, the viscosity of the resin can be adjusted to a low viscosity. Therefore, the thermosetting resin which has been adequately adjusted in viscosity can be easily impregnated into the chopped fiber bundle [A]. Further, by appropriately adjusting the viscosity of the thermosetting resin, even in any step for producing a fiber reinforced plastic, a resin can be added as needed. Further, the molding material, in which the resin is in a state of uncured condition at a room temperature, has a flexibility. Therefore, such a molding material is easy to be cut and to follow the shape of a mold, and is excellent in handling ability. Besides, since it is possible to design so as to impart tackiness at a room temperature, because such a molding material is integrated by merely pressing it with each other or to another base material, it becomes easy to laminate with each other or to another base material to form a laminated material.

**[0119]** As the matrix resin [M] used in the fiber-reinforced resin molding material, there is a case where a thermoplastic resin is preferred. Generally, since a thermoplastic resin has a high toughness, by using a thermoplastic resin as the matrix resin, it is possible to suppress the connection of cracks to each other, which is a weak point of a short fiber reinforced plastic, thereby improving the strength of the short fiber reinforced plastic. In particular, for applications that place importance on impact properties, it is better to use a thermoplastic resin for the matrix resin. Since molding using a thermoplastic resin usually does not accompany a chemical reaction, molding time can be shortened by using a thermosetting resin.

**[0120]** In the fiber-reinforced resin molding material of the present invention, the arrangement direction of the reinforcing fibers of each chopped fiber bundle [A] contained in the random mat may be the same. By laminating a plurality of molding materials having the same arrangement direction of reinforcing fibers, it becomes easy to design a laminate having desired physical properties. It becomes easy to reduce the variation of the mechanical properties of the obtained laminate. Such a molding material is produced by spraying a large number of chopped fiber bundles [A] used in the present invention onto a base material in the form of a sheet so that the arrangement directions of reinforcing fibers of each chopped fiber bundle [A] are the same. As means for spraying respective chopped fiber bundles [A] so that the arrangement directions of reinforcing fibers thereof are the same, for example, there is a slit-shaped nozzle capable of supplying the respective chopped fiber bundles [A] onto a base material at a state where the arrangement directions of reinforcing fibers are directed to a constant direction.

**[0121]** On the other hand, the fiber-reinforced resin molding material 191 in Fig. 20 is composed of an aggregate in which a large number of chopped fiber bundles [A] CFBs used in the present invention are positioned in a state of the reinforcing fibers thereof arranged in random directions. Although a large number of chopped fiber bundles [A] CFBs partially overlap with each other, no definite layer structure is formed. This fiber-reinforced resin molding material 191 can be produced inexpensively as compared with the case of producing a molding material having a layered structure while controlling the arrangement direction of reinforcing fibers of chopped fiber bundles [A], and further, it can be said that it is an isotropic and easy-to-design molding material.

**[0122]** The fiber-reinforced resin molding material of the present invention may be formed into a three-dimensional shape such that a random mat containing a large number of the above-described chopped fiber bundles [A] has at least

one bent portion in its cross-sectional shape. In case of molding a fiber reinforced plastic similarly having a three-dimensional shape using the fiber-reinforced resin molding material of the present invention having a three-dimensional shape, because it is not necessary to greatly flow the chopped fiber bundles [A] during molding, undulation and unevenness of the reinforcing fiber arrangement due to the flow are prevented, and an excellent quality stability can be obtained in an obtained molded article of the fiber reinforced plastic.

**[0123]** The fiber-reinforced resin molding material of the present invention having a three-dimensional shape can be produced, for example, as follows. Can be exemplified a method for producing a molding material comprising the steps of spraying a large number of chopped fiber bundles [A] used in the present invention and a matrix resin [M] onto a molding base material in a sheet-like form to integrate them, and forming the integrated sheet into a three-dimensional shape. For example, a method for producing a molding material comprising the steps of passing a large number of chopped fiber bundles [A] used in the present invention through a slit-shaped nozzle so that the arrangement directions of the reinforcing fibers of the respective chopped fiber bundles [A] are aligned in the same direction, spraying them onto a molding base material having a three-dimensional shape to form a layer comprising a chopped fiber bundle aggregate having the same arrangement directions of reinforcing fibers, and spraying a large number of the chopped fiber bundles [A] onto the previously formed layer in a sheet-like form so that the arrangement directions of the reinforcing fibers of the respective chopped fiber bundles [A] are aligned in different directions of the reinforcing fibers of the chopped fiber bundles [A] in the previously formed layer and so that the arrangement directions of the reinforcing fibers of the respective chopped fiber bundles [A] sprayed later are aligned in the same direction, can be exemplified.

**[0124]** Fig. 21 is a schematic perspective diagram for explaining an example of a method of producing a fiber-reinforced resin molding material of the present invention having a three-dimensional shape. In Fig. 21, an apparatus for producing a fiber-reinforced resin molding material of the present invention having a three-dimensional shape comprises, for example, a plurality of bobbins 202 each wound with a continuous reinforcing fiber bundle 201 (in Fig. 21, six bobbins are depicted), guide rollers 203 and 204 for the continuous reinforcing fibers, a roller cutter 205 for pulling out the continuous reinforcing fibers from the bobbins 202, passing the fiber bundle of the continuous reinforcing fibers pulled out through a partial separation processing device 211 for performing a partial separation processing to the fiber bundle, and thereafter, cutting the partially separated fiber bundle [B] 212 at constant intervals and in a direction inclined relatively to the lengthwise direction of the fiber bundle of the continuous reinforcing fibers, a sizing agent applying device 206 for applying a sizing agent to the chopped fiber bundle [A] obtained by cutting of the continuous reinforcing fibers, a sizing agent supplying port 207 provided on the side portion of the sizing agent applying device 206, a slit-shaped nozzle 208 for controlling the arrangement direction of the reinforcing fibers of the chopped fiber bundle [A] applied with the sizing agent to a predetermined direction, a molding base material 209 comprising a forming mold having a three-dimensional shape, and a robot arm 210.

**[0125]** The roller cutter 205 is attached to the upper portion of the sizing agent applying device 206. The sizing agent applying device 206 has a chopped fiber bundle introducing port, which receives the chopped fiber bundle [A] obtained by cutting, at its upper portion, and a chopped fiber bundle discharging port, which discharges the chopped fiber bundle [A] applied with the sizing agent, at its lower portion. The slit-shaped nozzle 208 has a chopped fiber bundle introducing port, which receives the chopped fiber bundle [A] discharged from the chopped fiber bundle discharging port, at its upper portion, and a chopped fiber bundle discharging port, which discharges the chopped fiber bundle [A] in which the arrangement directions of the reinforcing fibers are controlled to a constant direction, at its lower portion. The slit-shaped nozzle 208 is attached to the lower portion of the sizing agent applying device 206. The tip of the robot arm 210 is coupled to the side portion of the sizing agent applying device 206. The tip of the robot arm 210 is movable with respect to the molding base material 209 by a robot arm operating device (not shown).

**[0126]** In Fig. 21, by the rotation of the rollers of the roller cutter 205, the continuous reinforcing fiber bundle 201 pulled out from the bobbins 202 passes through the guide rollers 203 and 204, and passes through the partial separation processing device 211, and then, a partially separated fiber bundle [B] 212 is introduced into the roller cutter 205 where it is cut so as to form the chopped fiber bundle [A] used in the present invention. The chopped fiber bundles [A] obtained by cutting are introduced to the sizing agent applying device 206. Inside the sizing agent application device 206, the powdered sizing agent supplied from the sizing agent supplying port 207 is applied to the chopped fiber bundles [A]. The chopped fiber bundles [A] applied with the sizing agent are introduced into the slit-shaped nozzle 208. As the chopped fiber bundles moves inside the slit-shaped nozzle 208, the chopped fiber bundles [A] are aligned so that the arrangement direction of the reinforcing fibers becomes a constant direction. The aligned chopped fiber bundles [A] are discharged from the slit-shaped nozzle 208, drop while substantially being maintained with the arrangement state of the chopped fiber bundles, and reach the surface of the molding base material 209.

**[0127]** The arrival position on the surface of the molding base material 209 of the chopped fiber bundles is sequentially changed by the operation of the robot arm 210 and a layer of the chopped fiber bundles [A] adhered with the powdered sizing agent is formed on the molding base material 209. The layer of chopped fiber bundles [A] formed on the molding base material 209 is heated in order to melt the powdered sizing agent contained therein, and by the molten sizing agent, the chopped fiber bundles are integrated to each other, and the molding material of the present invention having a three-

dimensional shape is produced.

**[0128]** The fiber reinforced plastic molded using the fiber-reinforced resin molding material of the present invention contains the random mat containing the above-described chopped fiber bundles [A] and a matrix resin [M]. Therefore, in the chopped fiber bundle [A] in the fiber reinforced plastic of the present invention, the fiber length of the reinforcing fibers forming it is, for example, 5 to 100 mm, and the chopped fiber bundle [A] has transition sections in each of which the number of the reinforcing fibers in the cross section of the chopped fiber bundle [A] increases from each end of the chopped fiber bundle [A] along the arrangement direction of the reinforcing fibers toward the central portion in the lengthwise direction of the chopped fiber bundle [A], and over the entire range of the chopped fiber bundle [A], the change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the chopped fiber bundle [A] is 0.05 mm<sup>2</sup> or less per a length of 1 mm.

**[0129]** Since the chopped fiber bundle [A] in such a fiber reinforced plastic has a formation in which the number of reinforcing fibers decreases from the central portion thereof to the end portion, in the fiber reinforced plastic, the load received by the chopped fiber bundle [A] can be gradually delivered to the surrounding chopped fiber bundle [A], thereby effectively reducing the stress concentration. In particular, by the condition where the change amount of total cross-sectional area of reinforcing fibers is 0.05 mm<sup>2</sup> or less per 1 mm over the entire range of chopped fiber bundle [A], the stress transmission efficiency is drastically improved. This change amount is preferably 0.04 mm<sup>2</sup> or less. In order to minimize the influence of stress concentration, it is better that this change amount is 0.025 mm<sup>2</sup> or less. In case of carbon fibers having the diameter of a reinforcing fiber (a single fiber) of about 5 to 10 μm, it is better that the change amount of the total number of reinforcing fibers is 1,400 or less per 1 mm over the entire range of chopped fiber bundle [A]. This change amount of the total number is more preferably 1,000 or less. In order to minimize the influence of stress concentration, the change amount of the total number is preferably 800 or less.

#### Examples

**[0130]** Next, Examples and Comparative Examples of the present invention will be explained. Where, the present invention is not limited in any way by the Examples and Comparative Examples.

[Used material]

**[0131]** Fiber bundle [B-1]: A continuous carbon fiber bundle having a fiber diameter of 7.2 μm, a tensile elastic modulus of 240 GPa, and a number of single fibers of 50,000 ("Panex 35 (registered trademark)", supplied by ZOLTEK Co., Ltd.) was used.

**[0132]** Matrix resin [M-1]: A resin compound prepared by sufficiently mixing and stirring 100 parts by weight of a vinyl ester resin ("DELAKEN (registered trademark) 790", supplied by Dow Chemical Co., Ltd.), 1 part by weight of tert-butyl peroxybenzoate

**[0133]** ("Perbutyl (registered trademark) Z", supplied by NOF CORPORATION) as a curing agent, 4 parts by weight of magnesium oxide (MgO #40, supplied by Kyowa Chemical Industry Co., Ltd.) as a thickener, and 2 parts by weight of zinc stearate (SZ-2000, supplied by Sakai Chemical Industry Co., Ltd.) as an internal release agent.

[Classification of chopped fiber bundle [A] and calculation method of content of aggregate [Y]]

**[0134]** A sample of 100 mm × 100 mm was cut out from the fiber-reinforced resin molding material, and the sample was heated in a furnace at 600 °C for 1 hour to remove the resin. Subsequently, 400 chopped fiber bundles [A] were taken out from the sample from which the resin was removed using tweezers and they were classified into aggregate [X], aggregate [Y] and aggregate [Z] according to the following criteria.

aggregate [X]: In a partially separated fiber bundle, the small bundles divided originating from the applied separation processing were determined as separated bundle aggregates (aggregates [X]).

aggregate [Y]: In a partially separated fiber bundle, one capable of being judged to be "a shape joined with bundles with each other" by inter-bundle joining factors such as not-separation-processed section, entangled part, entanglement accumulation part, etc. was determined as joined bundle aggregate (aggregate [Y]).

aggregate [Z]: In a partially separated fiber bundle, one having a trace divided by cutting inter-bundle joining factors such as not-separation-processed section, entangled part, entanglement accumulation part, etc., or one capable of being judged to be made into a small piece by natural thread splitting in the process after being cut, was determined as joined cut aggregate (aggregate [Z]).

**[0135]** Furthermore, the content of aggregate [Y] in the fiber-reinforced resin molding material was calculated from the total number of aggregates [Y] classified as described above.

[Evaluation method of mechanical properties]

**[0136]** A mold No. 1 capable of producing a flat plate was used. A fiber-reinforced resin molding material was placed in the central portion of the mold No. 1 (50% in terms of charge rate), and then cured under a pressure of 10 MPa by a pressurizing-type press machine at a condition of about  $140^{\circ}\text{C} \times 5$  minutes to obtain a flat plate of  $300 \times 400$  mm. In a condition where the lengthwise direction of the flat plate was referred to as  $0^{\circ}$  direction, five pieces (total 10 pieces) of  $100 \times 25 \times 1.6$  mm specimens were cut out from the directions of  $0^{\circ}$  and  $90^{\circ}$ , respectively, from the obtained flat plate, and the measurement was carried out in accordance with JIS K 7074 (1988) (flexural strength [MPa], flexural modulus [GPa], CV (coefficient of variation) of flexural modulus [%]).

(Example 1)

**[0137]** The fiber bundle [B-1] was unwound at a constant speed of 10 m/min using a winder, passed through a vibration widening roll vibrating in its axial direction at 10 Hz, and after a widening treatment was applied, it was passed through a width regulating roll having a width of 60 mm to obtain a widened fiber bundle widened to 60 mm in width. For the obtained widened fiber bundle, a separation processing means was prepared in which iron plates for separation processing each having a protruding shape with a thickness of 0.2 mm, a width of 3 mm and a height of 20 mm were arranged in parallel to each other at equal intervals of 3.5 mm with respect to the width direction of the reinforcing fiber bundle. This separation processing means was intermittently pierced into and pulled out from the widened fiber bundle to obtain a partially separated fiber bundle. At this time, the separation processing means was pierced into the widened fiber bundle running at a constant speed of 10 m/min for 3 seconds to create a separation-processed section, pulled out therefrom in 0.2 second, and the operation of piercing again was repeated.

**[0138]** In the partially separated fiber bundle obtained, the fiber bundle was divided into 17 parts in the width direction in the separation-processed section, and in at least one end portion of at least one separation-processed section, an entanglement accumulation part, in which entangled parts each tangled with single fibers were accumulated, was present. When the partially separated fiber bundle was manufactured by 1,500 m, the twists of the fibers existing in the fiber bundle passed through in the running direction when pulling out and piercing the separation processing means without causing yarn breakage and winding at all, and the separation processing could be carried out with the stable width.

**[0139]** The obtained partially separated fiber bundle was inserted continuously into a rotary cutter whose cutting blade was inclined at an angle of  $15^{\circ}$  with respect to the lengthwise direction of the fiber bundle to cut the fiber bundle, and the chopped fiber bundle [A] was obtained. At this time, the cutting interval was adjusted to 6.5 mm in advance so that it could be cut to a fiber length of 25 mm. Further, although the inserted partially separated fiber bundle was widened up to 60 mm width when applying the above-described separation processing process due to the winding process of the partially separated fiber bundle and thread tension applied during cutting process, the width W of the fiber bundle at the time of cutting was 20 mm. The obtained chopped fiber bundle [A] had a shape having an unchanging section 114 and two transition sections 113a and 113b as shown in Fig. 15. At this time, although the change amount of the total cross-sectional area of the reinforcing fibers in the transition section 113a and 113b varies depending on the classification of the form of the chopped fiber bundle [A], it was 0.005 to 0.025 mm<sup>2</sup> per 1 mm in the lengthwise direction of the chopped fiber bundle.

**[0140]** Following the cutting process described above, the chopped fiber bundles [A] were sprayed so as to be uniformly dispersed to obtain a random mat whose fiber orientation is isotropic. The areal weight of the obtained discontinuous fiber nonwoven fabric was 1 kg/m<sup>2</sup>.

**[0141]** The matrix resin [M-1] was uniformly applied to respective two sheets of polypropylene release films using a doctor blade to make two resin sheets. A sheet-like fiber-reinforced resin molding material was obtained by nipping the above-described obtained random mat with these two resin sheets and impregnating the resin into the mat by a roller. At this time, the amount of resin application was adjusted at the stage of manufacturing the resin sheets so that the weight content of the reinforcing fibers of the fiber-reinforced resin molding material became 47%.

**[0142]** With respect to the obtained fiber-reinforced resin molding material, when the content of the aggregates [Y] was calculated based on the classification of the chopped fiber bundle [A] and the method of calculating the content of the aggregate [Y], it was 10%. Further, based on the aforementioned evaluation method of mechanical properties, a fiber-reinforced resin molding material was molded and the mechanical properties were evaluated. A series of evaluation results obtained are shown in Table 1.

(Example 2)

**[0143]** The evaluations were carried out in the same manner as in Example 1 except that the cutting interval was adjusted to 3.2 mm so that the fiber length of the bundled aggregate [A] was 12.5 mm. A series of evaluation results obtained are shown in Table 1.

(Example 3)

[0144] The evaluations were carried out in the same manner as in Example 1 except that the inclination of the cutting blade of the rotary cutter was adjusted and the cutting interval was adjusted to 6.2 mm so that the cutting angle of the fiber bundle became 30° and the fiber length became 12.5 mm. A series of evaluation results obtained are shown in Table 1.

(Example 4)

[0145] The evaluations were carried out in the same manner as in Example 1 except that the width of the partially separated fiber bundle was adjusted so that the width W of the partially separated fiber bundle at the time of cutting became 30 mm, by providing a squeezing roller for maintaining the widened width of the fiber bundle immediately before the winding of the partially separated fiber bundle 1. A series of evaluation results obtained are shown in Table 1.

(Comparative Example 1)

[0146] The evaluations were carried out in the same manner as in Example 1 except that a chopped fiber bundle [A] was obtained by using a rotary cutter with cutting blades installed at an angle of 90° and a cutting interval of 25 mm with respect to the lengthwise direction of the fiber bundle when cutting the partially separated fiber bundle. A series of evaluation results obtained are shown in Table 2.

(Comparative Example 2)

[0147] The evaluations were carried out in the same manner as in Example 1 except that a chopped fiber bundle [A] was obtained by cutting the fiber bundle [B-1] without performing a separation processing thereto. A series of evaluation results obtained are shown in Table 2.

(Table 1)

		Unit	Example			
			1	2	3	4
Used raw material	Fiber bundle	-	[B-1]	[B-1]	[B-1]	[B-1]
	Matrix resin	-	[M-1]	[M-1]	[M-1]	[M-1]
Bundled aggregate [A]	Cutting angle $\theta$	°	15	15	30	15
	Fiber length	mm	25	12.5	12.5	12.5
	Width of fiber bundle at cutting W	mm	20	20	20	30
	Value of $W \cdot \cos \theta / D$	-	3	6	3	9
	Change amount of total cross-sectional area in transition section	mm <sup>2</sup> /mm	0.005 - 0.025	0.005 - 0.025	0.023 - 0.047	0.011 - 0.020
	Content of aggregates [Y]	%	10	9	12	8
Mechanical properties	Flexural strength	MPa	415	435	385	450
	Flexural modulus	GPa	25	26	23	27
	CV of flexural modulus	%	13	10	16	8

(Table 2)

		Unit	Comparative Example		
			1	2	
5	Used raw material	Reinforcing fiber	-	[B-1]	[B-1]
		Matrix resin	-	[M-1]	[M-1]
10	Bundled aggregate [A]	Cutting angle $\theta$	$^{\circ}$	90	15
		Fiber length	mm	25	25
		Width of fiber bundle at cutting W	mm	20	20
		Value of $W \cdot \cos \theta / D$	-	0	3
		Change amount of total cross-sectional area in transition section	mm <sup>2</sup> /mm	-	0.027
		Content of aggregates [Y]	%	30	45
15	Mechanical properties	Flexural strength	MPa	320	385
		Flexural modulus	GPa	20	24
		CV of flexural modulus	%	27	24

25 **[0148]** In Examples 1 to 4, it was confirmed that excellent mechanical properties (flexural strength, elastic modulus) and low variation were both exhibited. In Example 3, although it was observed that the mechanical properties were reduced because the stress concentration at the end portion of the fiber bundle was increased by increasing the cutting angle, it was confirmed to be no problem in level. Further, in Example 4, it was confirmed that by adjusting the width of the fiber bundle at the time of cutting, inter-bundle coupling factors such as not-separation-processed section, entangled part, and entanglement accumulation part can be subdivided, and it had a remarkable effect on improvement of mechanical properties and reduction in variation.

30 **[0149]** On the other hand, with respect to Comparative Examples 1 and 2, in Comparative Example 1, since the fiber bundle was cut at a cutting angle of 90°, stress concentration occurred at the end portion of the fiber bundle, and further, because the content of aggregates [Y] was high, a decrease in mechanical properties and an increase in variation were observed. Further, in Comparative Example 2, since the separation processing was not performed to the reinforcing fiber bundle, the content of aggregates [Y] was high, a decrease in mechanical properties and an increase in variation were observed.

#### Industrial Applicability

40 **[0150]** The present invention is applicable to any fiber-reinforced resin molding material which is required to have excellent flowability at the time of molding, high mechanical properties of the molded article and reduction of the variation of its mechanical properties.

#### Explanation of symbols

45

#### **[0151]**

- 50 1: 1, 17, 31, 41, 51, 61, 71, 81, 91, 212: partially separated fiber bundle [B]  
 2, 13, 15, 23, 32, 42, 64, 74: separation-processed section  
 3, 14, 16, 28, 33, 52, 62, 72, 82: not-separation-processed section  
 4: cutting blade  
 5: chopped fiber bundle [A]  
 11, 25, 63: entangled part  
 12, 26, 73: entanglement accumulation part  
 55 20: fiber bundle  
 21: separation means  
 22: protrusion

24: contact portion  
 27: fluff accumulation  
 34, 35, 43, 53, 65, 75, 83, 92: cutting plane  
 36, 37: bundled aggregate  
 5 F, 111: reinforcing fiber  
 112a: first tip  
 112b: second tip  
 113a: first transition section  
 113b: second transition section  
 10 113Ea: first terminal end surface of transition section  
 113Eb: second terminal end surface of transition section  
 114: unchanging section  
 114Ea: one end surface of unchanging section  
 115a: one side of first transition section  
 15 115b: one side of second transition section  
 116a: the other side of first transition section  
 116b: the other side of second transition section  
 191: fiber-reinforced resin molding material  
 201: continuous reinforcing fiber bundle  
 20 202: bobbin  
 203, 204: guide roller  
 205: roller cutter  
 206: sizing agent applying device  
 207: sizing agent supplying port  
 25 208: slit-shaped nozzle  
 209: molding base material  
 210: robot arm  
 211: partial separation processing device  
 $\theta$ : cutting angle  
 30 L: lengthwise direction  
 CFB, CFB1, CFB2, CFB3, CFB5a - CFB5g: chopped fiber bundle [A]  
 CFTS: change amount of total cross-sectional area of reinforcing fibers  
 Ld: passing length of chopped fiber bundle [A]  
 Lf: fiber length of reinforcing fiber  
 35 Tb: maximum thickness of chopped fiber bundle [A]  
 Wb: maximum width of chopped fiber bundle [A]  
 Wd: passing width of chopped fiber bundle [A]

40 **Claims**

1. A random mat comprising at least a chopped fiber bundle [A] (5), wherein the chopped fiber bundle [A] (5) satisfies at least the following (a) to (d):

45 (a) the chopped fiber bundle [A] (5) is a bundled aggregate of discontinuous reinforcing fibers obtained by cutting a partially separated fiber bundle [B] (91) prepared by alternately forming separation-processed sections (2), each of which is separated into a plurality of bundles, and not-separation-processed sections (3), along a lengthwise direction (L) of a fiber bundle comprising a plurality of reinforcing fibers (111);  
 50 (b) the chopped fiber bundle [A] has a first transition section (113a) in which the number of the reinforcing fibers (111) in a cross section of the fiber bundle in a direction perpendicular to the lengthwise direction (L) of the fiber bundle is increased from a first tip (112a) which is one tip in the lengthwise direction (L) of the fiber bundle, toward a second tip (112b) which is the other tip, and a second transition section (113b) in which the number of the reinforcing fibers (111) in the cross section of the fiber bundle is increased from the second tip (112b) toward the first tip (112a);  
 55 (c) between the first transition section (113a) and the second transition section (113b), along the lengthwise direction (L) of the fiber bundle, an unchanging section (114) in which the number of the reinforcing fibers (111) in the cross section of the fiber bundle is invariable is provided, and one end surface (114Ea) of the unchanging section (114) coincides with a first terminal end surface (113Ea) which is a terminal end at a side opposite to

the first tip (112a) of the first transition section (113a) and the other end surface (114Eb) of the unchanging section (114) coincides with a second terminal end surface (113Eb) which is a terminal end at a side opposite to the second tip (112b) of the second transition section (113b), or the first terminal end surface (113Ea) and the second terminal end surface (113Eb) coincide directly with each other; and

(d) a change amount of the total cross-sectional area of the reinforcing fibers in the cross section of the fiber bundle between the first tip (112a) and the second tip (112b) is 0.05mm<sup>2</sup> or less per a length of 1 mm in the lengthwise direction (L) of the fiber bundle,

wherein the partially separated fiber bundle [B] (91) is cut at an angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) with respect to the lengthwise direction (L) of the fiber bundle when obtaining the chopped fiber bundle [A] (1).

2. The random mat according to claim 1, wherein in the partially separated fiber bundle [B] (91), an entangled part in which the reinforcing fibers are entangled at least at one end of at least one of the separation-processed sections, and/or an entanglement accumulation part (12; 26; 73) in which the entangled parts are accumulated, is formed.

3. The random mat according to claim 1 or 2, wherein bundled aggregates of the discontinuous reinforcing fibers obtained by cutting the partially separated fiber bundle [B] (91) are classified into at least the following aggregates [X] to [Z], and the chopped fiber bundle [A] comprises at least one of the aggregates [X], [Y] and [Z]:

aggregate [X]: a separated bundle aggregate divided into arbitrary number of bundles by separation processing; aggregate [Y]: a joined bundle aggregate in which the reinforcing fibers of fiber bundles are joined to each other by being formed with the not-separation-processed section, and/or an entangled part (11; 25; 63) in which the reinforcing fibers are entangled at least at one end of at least one of the separation-processed sections, and/or an entanglement accumulation part (12; 26; 73) in which the entangled parts are accumulated; and aggregate [Z]: a joined cut aggregate in which a cutting plane at cutting of the partially separated fiber bundle is intersected with the not-separation-processed section and/or the entangled part (11; 25; 63) and/or the entanglement accumulation part (12; 26; 73), and at an intersected part, the joining of the reinforcing fibers of fiber bundles to each other is cut.

4. The random mat according to claim 3, wherein the content of the joined bundle aggregate [Y] in the bundled aggregates of the discontinuous reinforcing fibers obtained by cutting the partially separated fiber bundle [B] (91) is in the range of 0 to 15%.

5. A method for producing a random mat according to any one of claims 1 to 4, wherein the partially separated fiber bundle [B] (91) is cut at an angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) with respect to the lengthwise direction of the fiber bundle when obtaining the chopped fiber bundle [A] (5).

6. The method according to claim 5, wherein the partially separated fiber bundle [B] (91) is cut so as to satisfy the following equation (1) when obtaining the chopped fiber bundle [A] (5):

$$W \cdot \cos \theta / D \geq 3 \cdots (1)$$

W: width of fiber bundle when cutting partially separated fiber bundle

D: distance between cutting planes in chopped fiber bundle [A]

7. A fiber-reinforced resin molding material (191) comprising a random mat according to any one of claims 1 to 4 and a matrix resin [M].

**Patentansprüche**

1. Wirrvlies, umfassend mindestens ein geschnittenes Faserbündel [A] (5), wobei das geschnittene Faserbündel [A] (5) zumindest die folgenden Punkte (a) bis (d) erfüllt:

(a) das geschnittene Faserbündel [A] (5) ist ein gebündeltes Aggregat diskontinuierlicher Verstärkungsfasern, das durch Schneiden eines teilweise getrennten Faserbündels [B] (91), das durch abwechselndes Bilden von trennungsverarbeiteten Abschnitten (2), von denen jeder in eine Vielzahl von Bündeln getrennt ist, und nicht

trennungsverarbeiteten Abschnitten (3) hergestellt wird, entlang einer Längsrichtung (L) eines Faserbündels, das eine Vielzahl von Verstärkungsfasern (111) umfasst, erhalten wird;

(b) das geschnittene Faserbündel [A] weist einen ersten Übergangsabschnitt (113a), in dem die Anzahl der Verstärkungsfasern (111) in einem Querschnitt des Faserbündels in einer Richtung senkrecht zur Längsrichtung (L) des Faserbündels von einer ersten Spitze (112a), die eine Spitze in der Längsrichtung (L) des Faserbündels ist, in Richtung einer zweiten Spitze (112b), die die weitere Spitze ist, und einen zweiten Übergangsabschnitt (113b) auf, in dem die Anzahl der Verstärkungsfasern (111) im Querschnitt des Faserbündels von der zweiten Spitze (112b) zur ersten Spitze (112a) zunimmt;

(c) zwischen dem ersten Übergangsabschnitt (113a) und dem zweiten Übergangsabschnitt (113b) ist entlang der Längsrichtung (L) des Faserbündels ein unveränderlicher Abschnitt (114) vorgesehen, in dem die Anzahl der Verstärkungsfasern (111) im Querschnitt des Faserbündels unveränderlich ist, und eine Endfläche (114Ea) des unveränderlichen Abschnitts (114) fällt mit einer ersten Abschlussendfläche (113Ea) zusammen, die ein Abschlussende an einer Seite gegenüber der ersten Spitze (112a) des ersten Übergangsabschnitts (113a) ist, und die andere Endfläche (114Eb) des unveränderlichen Abschnitts (114) fällt mit einer zweiten Abschlussendfläche (113Eb) zusammen, die ein Abschlussende an einer Seite gegenüber der zweiten Spitze (112b) des zweiten Übergangsabschnitts (113b) ist, oder die erste Endfläche (113Ea) und die zweite Endfläche (113Eb) fallen direkt miteinander zusammen; und

(d) ein Änderungsbetrag der Gesamtquerschnittsfläche der Verstärkungsfasern im Querschnitt des Faserbündels zwischen der ersten Spitze (112a) und der zweiten Spitze (112b) beträgt  $0,05 \text{ mm}^2$  oder weniger pro Länge von 1 mm in der Längsrichtung (L) des Faserbündels,

wobei das teilweise getrennte Faserbündel [B] (91) unter einem Winkel  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) in Bezug auf die Längsrichtung (L) des Faserbündels beim Erhalten des geschnittenen Faserbündels [A] (1) geschnitten wird.

2. Wirrvlies nach Anspruch 1, wobei im teilweise getrennten Faserbündel [B] (91) ein Verschlingungsabschnitt, in dem die Verstärkungsfasern an zumindest einem Ende von zumindest einem der trennungsverarbeiteten Abschnitte verschlungen sind, und/oder ein Verschlingungsanhäufungsabschnitt (12; 26; 73) gebildet ist, in dem die Verschlingungsabschnitte angehäuft sind.

3. Wirrvlies nach Anspruch 1 oder 2, wobei gebündelte Aggregate der diskontinuierlichen Verstärkungsfasern, die durch Schneiden des teilweise getrennten Faserbündels [B] (91) erhalten werden, in zumindest die folgenden Aggregate [X] bis [Z] klassifiziert werden, und das geschnittene Faserbündel [A] zumindest eines der Aggregate [X], [Y] und [Z] umfasst:

Aggregat [X]: ein getrenntes Bündelaggregat, das durch einen Trennungsprozess in eine beliebige Anzahl von Bündeln aufgeteilt wurde;

Aggregat [Y]: ein verbundenes Bündelaggregat, in dem die Verstärkungsfasern von Faserbündeln miteinander dadurch verbunden sind, dass diese mit dem nicht trennungsverarbeiteten Abschnitt gebildet sind, und/oder ein verschlungener Abschnitt (11; 25; 63), bei dem die Verstärkungsfasern an zumindest einem Ende von zumindest einer der trennungsverarbeiteten Abschnitte verschlungen sind, und/oder ein Verschlingungsanhäufungsabschnitt (12; 26; 73), in dem die verschlungenen Abschnitte angehäuft sind; und

Aggregat [Z]: ein verbundenes geschnittenes Aggregat, bei dem eine Schnittebene beim Schneiden des teilweise getrennten Faserbündels mit dem nicht trennungsverarbeiteten Abschnitt und/oder dem verschlungenen Abschnitt (11; 25; 63) und/oder dem Verschlingungsanhäufungsabschnitt (12; 26; 73) geschnitten wird, und an einem geschnittenen Abschnitt die Verbindung der Verstärkungsfasern von Faserbündeln miteinander geschnitten wird.

4. Wirrvlies nach Anspruch 3, wobei der Gehalt des verbundenen Bündelaggregats [Y] in den gebündelten Aggregaten der diskontinuierlichen Verstärkungsfasern, die durch Schneiden des teilweise getrennten Faserbündels [B] (91) erhalten werden, im Bereich von 0 bis 15 % liegt.

5. Verfahren zum Herstellen eines Wirrvlieses nach einem der Ansprüche 1 bis 4, wobei das teilweise getrennte Faserbündel [B] (91) unter einem Winkel  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) in Bezug auf die Längsrichtung (L) des Faserbündels beim Erhalten des geschnittenen Faserbündels [A] (5) geschnitten wird.

6. Verfahren nach Anspruch 5, wobei das teilweise getrennte Faserbündel [B] (91) so geschnitten wird, dass die folgende Gleichung (1) beim Erhalten des geschnittenen Faserbündels [A] (5) erfüllt wird:

$$W \cos \theta / D \leq 3 \dots (1)$$

W: Breite des Faserbündels beim Schneiden eines teilweise getrennten Faserbündels

D: Abstand zwischen Schnittebenen im geschnittenen Faserbündel [A].

7. Faserverstärktes Harzformmaterial (191), umfassend ein Wirrvlies nach einem der Ansprüche 1 bis 4 und ein Matrixharz [M].

## Revendications

1. Mat aléatoire comprenant au moins un faisceau de fibres coupées [A] (5), dans lequel le faisceau de fibres coupées [A] (5) satisfait au moins les points (a) à (d) suivants :

(a) le faisceau de fibres coupées [A] (5) est un agrégat en faisceau de fibres de renforcement discontinues obtenu en coupant un faisceau de fibres partiellement séparé [B] (91) préparé en formant de manière alternée des sections traitées par séparation (2), dont chacune est séparée en une pluralité de faisceaux, et des sections non traitées par séparation (3), le long d'une direction de la longueur (L) d'un faisceau de fibres comprenant une pluralité de fibres de renforcement (111) ;

(b) le faisceau de fibres coupées [A] a une première section de transition (113a) dans laquelle le nombre de fibres de renforcement (111) dans une section transversale du faisceau de fibres dans une direction perpendiculaire à la direction de la longueur (L) du faisceau de fibres augmente d'une première pointe (112a) qui est une pointe dans la direction de la longueur (L) du faisceau de fibres, vers une deuxième pointe (112b) qui est l'autre pointe, et une deuxième section de transition (113b) dans laquelle le nombre de fibres de renforcement (111) dans la section transversale du faisceau de fibres augmente de la deuxième pointe (112b) vers la première pointe (112a) ;

(c) entre la première section de transition (113a) et la deuxième section de transition (113b), le long de la direction de la longueur (L) du faisceau de fibres, une section inchangée (114) dans laquelle le nombre de fibres de renforcement (111) dans la section transversale du faisceau de fibres est invariable est prévue, et une surface d'extrémité (114Ea) de la section inchangée (114) coïncide avec une première surface d'extrémité terminale (113Ea) qui est une extrémité terminale au niveau d'un côté opposé à la première pointe (112a) de la première section de transition (113a) et l'autre surface d'extrémité (114Eb) de la section inchangée (114) coïncide avec une deuxième surface d'extrémité terminale (113Eb) qui est une extrémité terminale au niveau d'un côté opposé à la deuxième pointe (112b) de la deuxième section de transition (113b), ou la première surface d'extrémité terminale (113Ea) et la deuxième surface d'extrémité terminale (113Eb) coïncident directement l'une avec l'autre ; et

(d) une quantité de changement de la surface de section transversale totale des fibres de renforcement dans la section transversale du faisceau de fibres entre la première pointe (112a) et la deuxième pointe (112b) est inférieure ou égale à 0,05 mm<sup>2</sup> pour une longueur de 1 mm dans la direction de la longueur (L) du faisceau de fibres,

dans lequel le faisceau de fibres partiellement séparé [B] (91) est coupé selon un angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) par rapport à la direction de la longueur (L) du faisceau de fibres lors de l'obtention du faisceau de fibres coupées [A] (1).

2. Mat aléatoire selon la revendication 1, dans lequel, dans le faisceau de fibres partiellement séparé [B] (91), une partie enchevêtrée dans laquelle les fibres de renforcement sont enchevêtrées au moins au niveau d'une extrémité d'au moins l'une des sections traitées par séparation, et/ou une partie d'accumulation d'enchevêtrement (12 ; 26 ; 73) dans laquelle les parties enchevêtrées sont accumulées, est/sont formée(s).

3. Mat aléatoire selon la revendication 1 ou 2, dans lequel les agrégats en faisceaux des fibres de renforcement discontinues obtenus en coupant le faisceau de fibres partiellement séparé [B] (91) sont classés en au moins les agrégats [X] à [Z] suivants, et le faisceau de fibres coupées [A] comprend au moins l'un des agrégats [X], [Y] et [Z] :

agrégat [X]: un agrégat de faisceau séparé divisé en un nombre arbitraire de faisceaux par traitement de séparation ;

agrégat [Y]: un agrégat de faisceau relié dans lequel les fibres de renforcement des faisceaux de fibres sont reliées les unes aux autres en étant formées avec la section non traitée par séparation, et/ou une partie en-

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chevêtrée (11 ; 25 ; 63) dans laquelle les fibres de renforcement sont enchevêtrées au moins au niveau d'une extrémité d'au moins l'une des sections traitées par séparation, et/ou une partie d'accumulation d'enchevêtrement (12 ; 26 ; 73) dans laquelle les parties enchevêtrées sont accumulées ; et

5 agrégat [Z] : un agrégat coupé relié dans lequel un plan de coupe à la coupe du faisceau de fibres partiellement séparé est traversé par la section non traitée par séparation et/ou la partie enchevêtrée (11 ; 25 ; 63) et/ou la partie d'accumulation d'enchevêtrement (12 ; 26 ; 73), et au niveau d'une partie d'intersection, la liaison des fibres de renforcement des faisceaux de fibres les unes aux autres est coupée.

10 4. Mat aléatoire selon la revendication 3, dans lequel la teneur de l'agrégat de faisceau relié [Y] dans les agrégats en faisceaux des fibres de renforcement discontinues obtenus en coupant le faisceau de fibres partiellement séparé [B] (91) se trouve dans la plage allant de 0 à 15%.

15 5. Procédé de production d'un mat aléatoire selon l'une quelconque des revendications 1 à 4, dans lequel le faisceau de fibres partiellement séparé [B] (91) est coupé selon un angle  $\theta$  ( $4^\circ \leq \theta \leq 25^\circ$ ) par rapport à la direction de la longueur du faisceau de fibres lors de l'obtention du faisceau de fibres coupées [A] (5).

20 6. Procédé selon la revendication 5, dans lequel le faisceau de fibres partiellement séparé [B] (91) est coupé de manière à satisfaire l'équation (1) suivante lors de l'obtention du faisceau de fibres coupées [A] (5) :

$$W \cdot \cos \theta / D \geq 3 \dots (1)$$

25 W: la largeur du faisceau de fibres lors de la coupe d'un faisceau de fibres partiellement séparé

D: la distance entre les plans de coupe dans le faisceau de fibres coupées [A].

30 7. Matériau de moulage en résine renforcée de fibres (191) comprenant un mat aléatoire selon l'une quelconque des revendications 1 à 4 et une résine matricielle [M].

FIG. 1

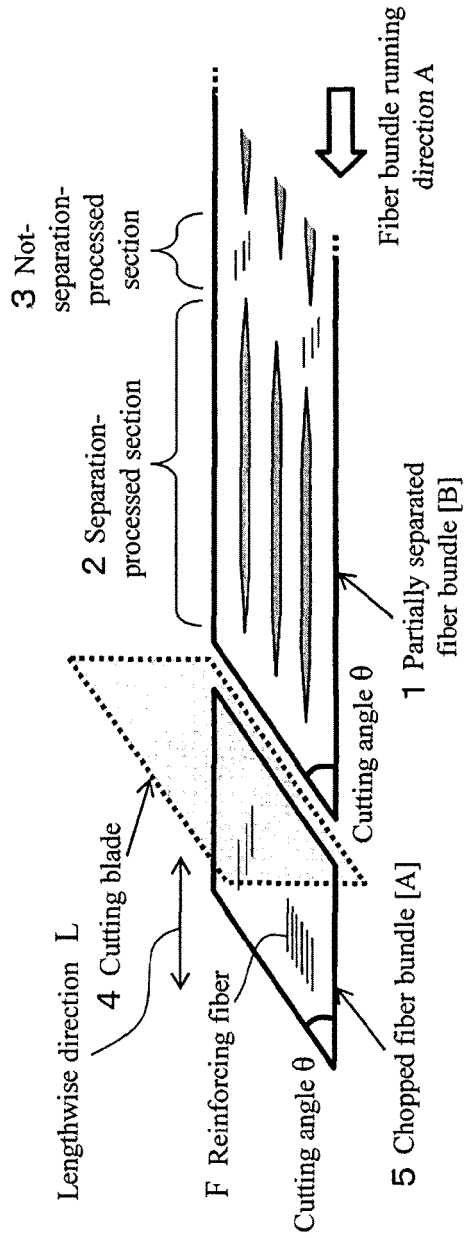
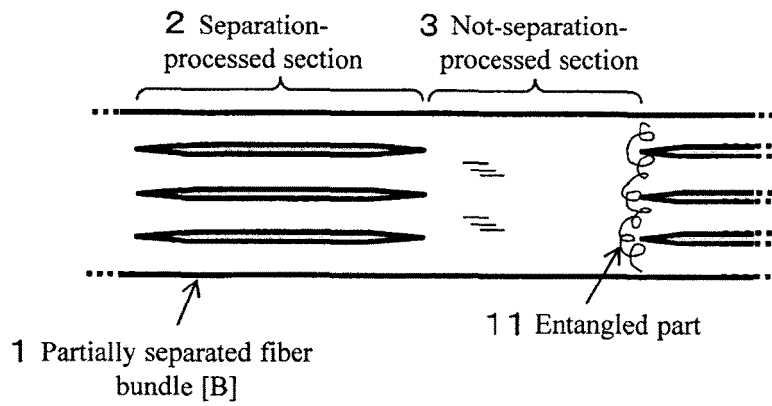
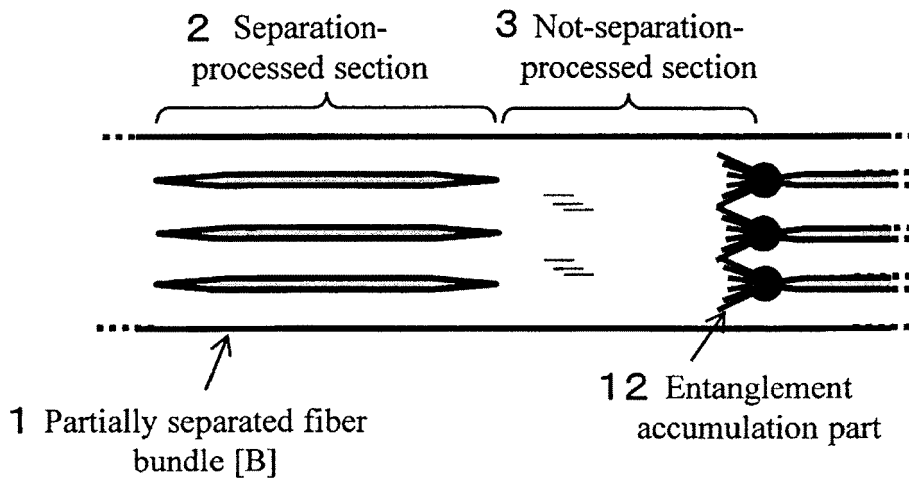


FIG. 2



**FIG. 3**



**FIG. 4**

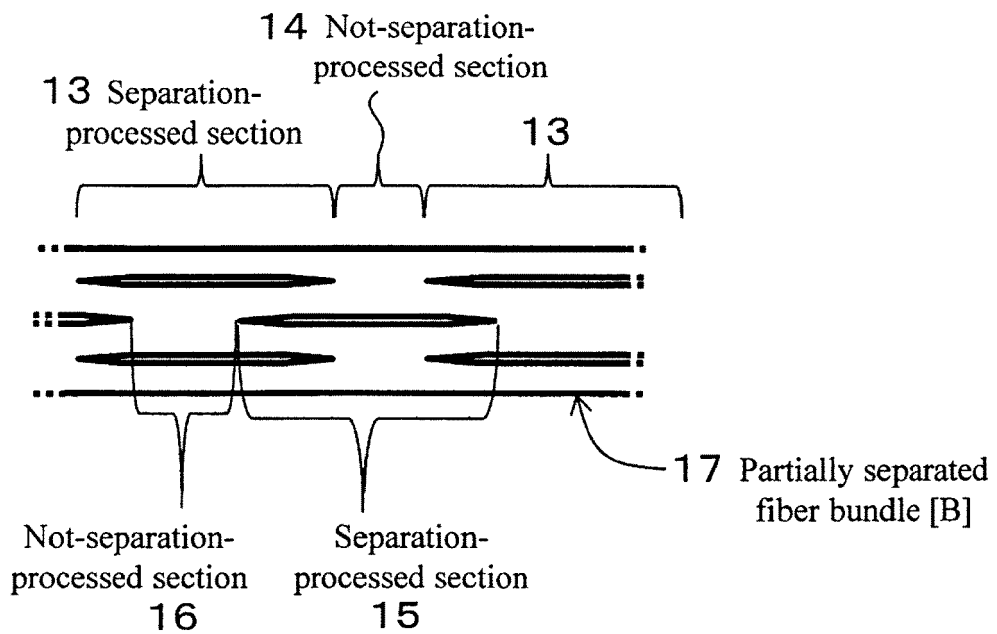


FIG. 5

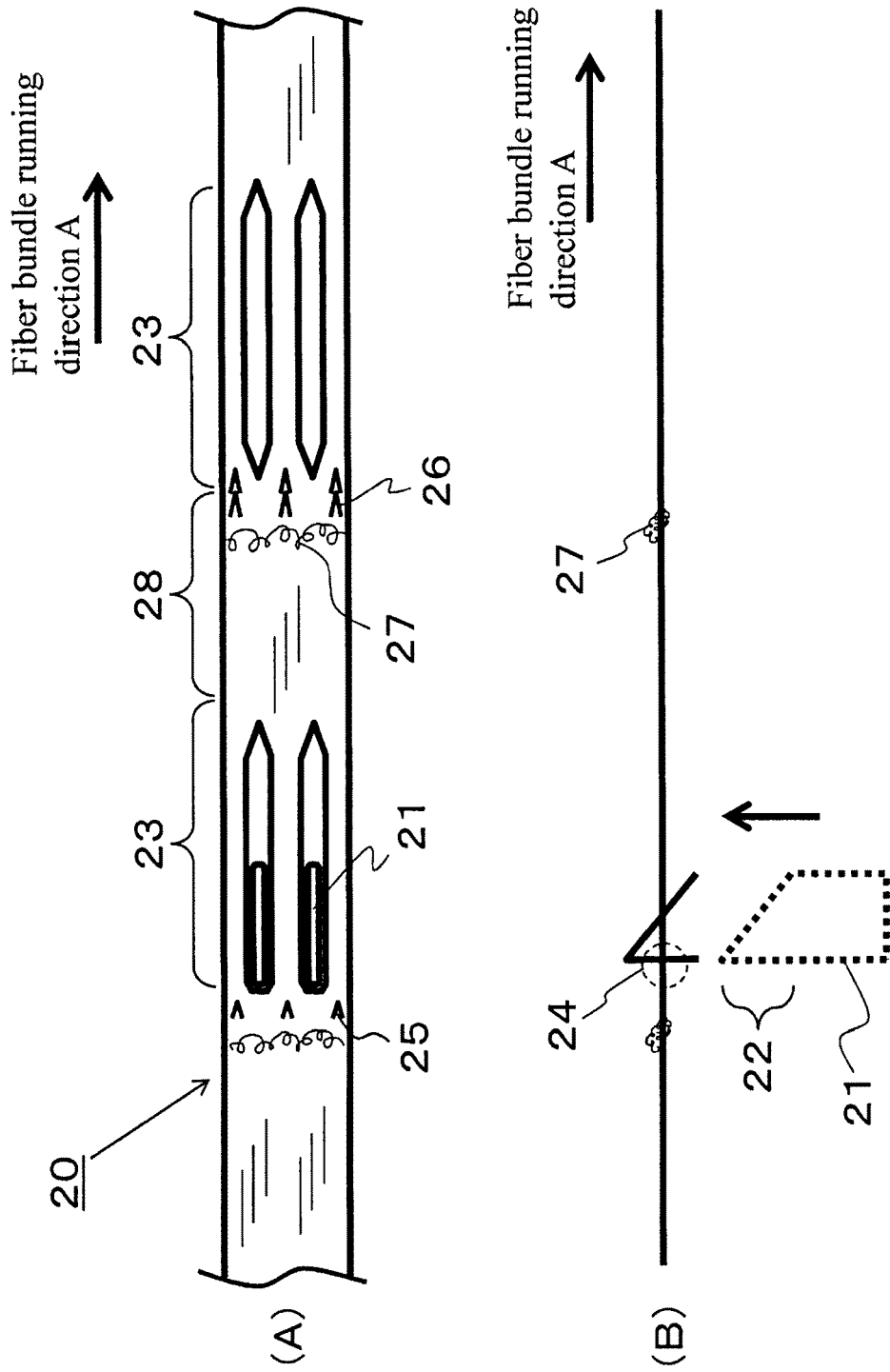
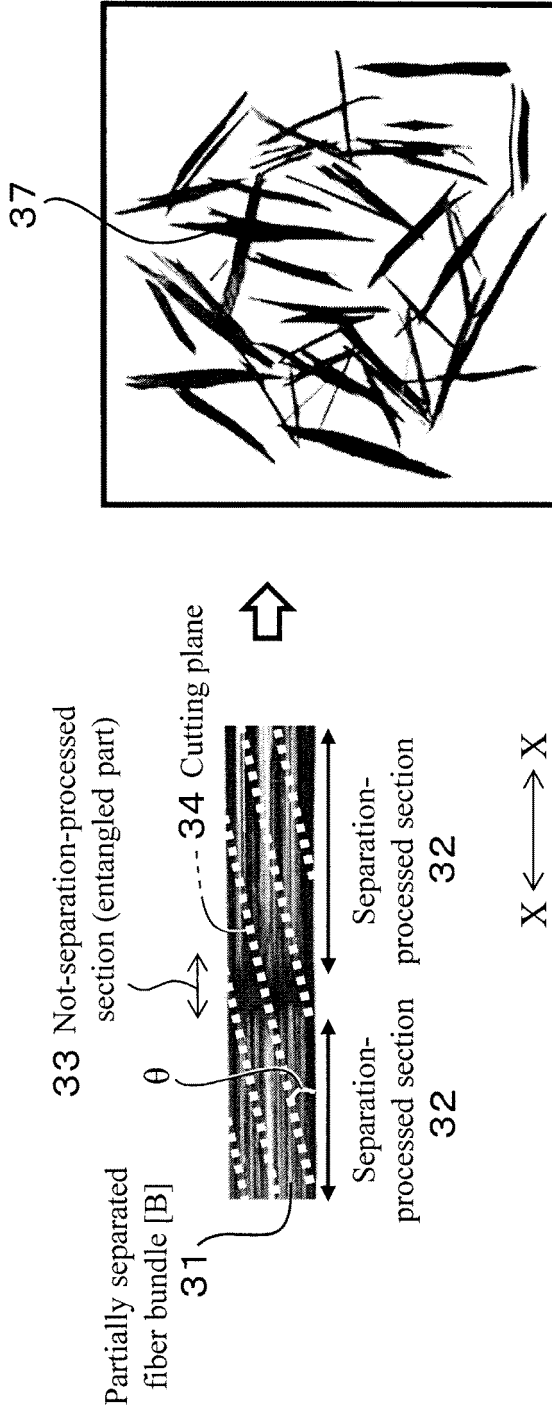
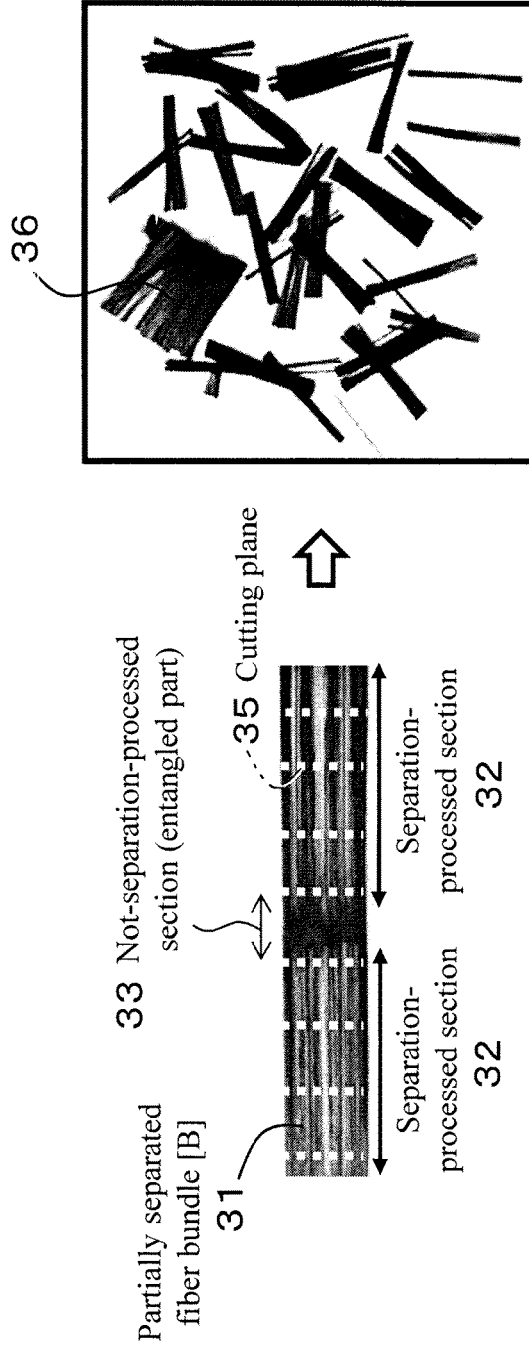


FIG. 6



Distribution diagram of discontinuous reinforcing fiber bundled aggregate

FIG. 7



Distribution diagram of discontinuous reinforcing fiber bundled aggregate

X ← → X

FIG. 8

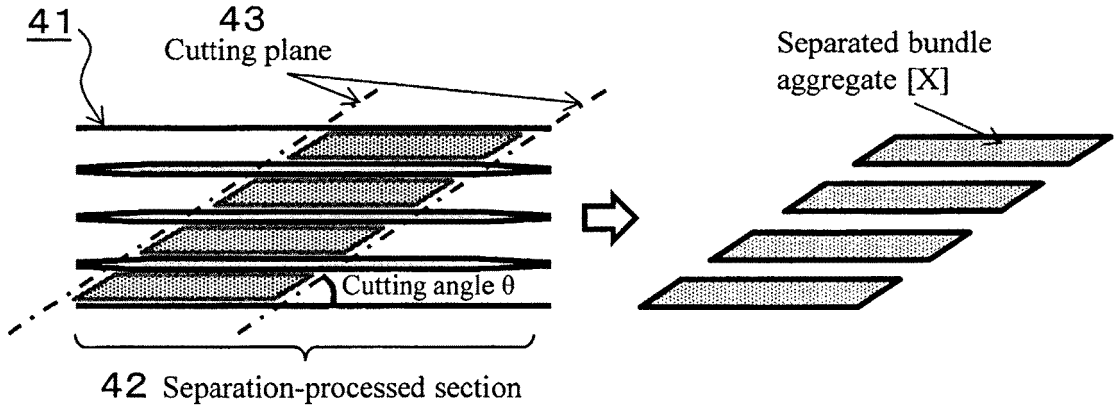


FIG. 9

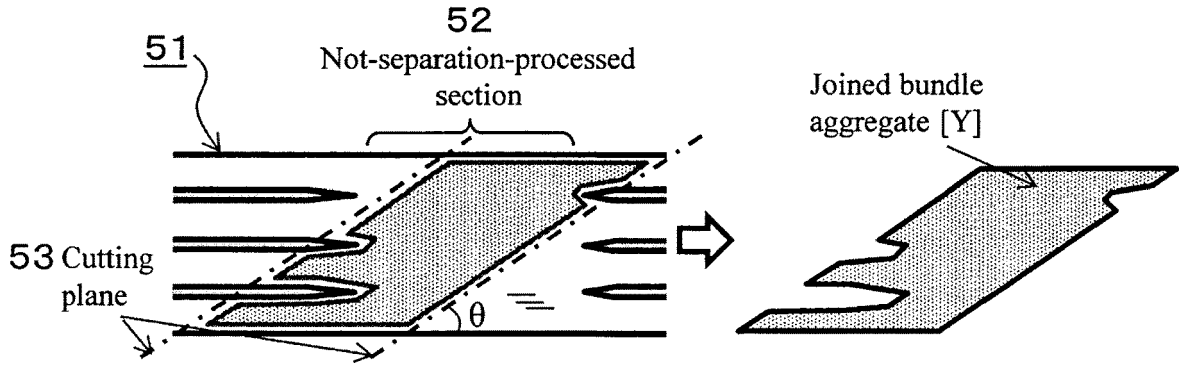


FIG. 10

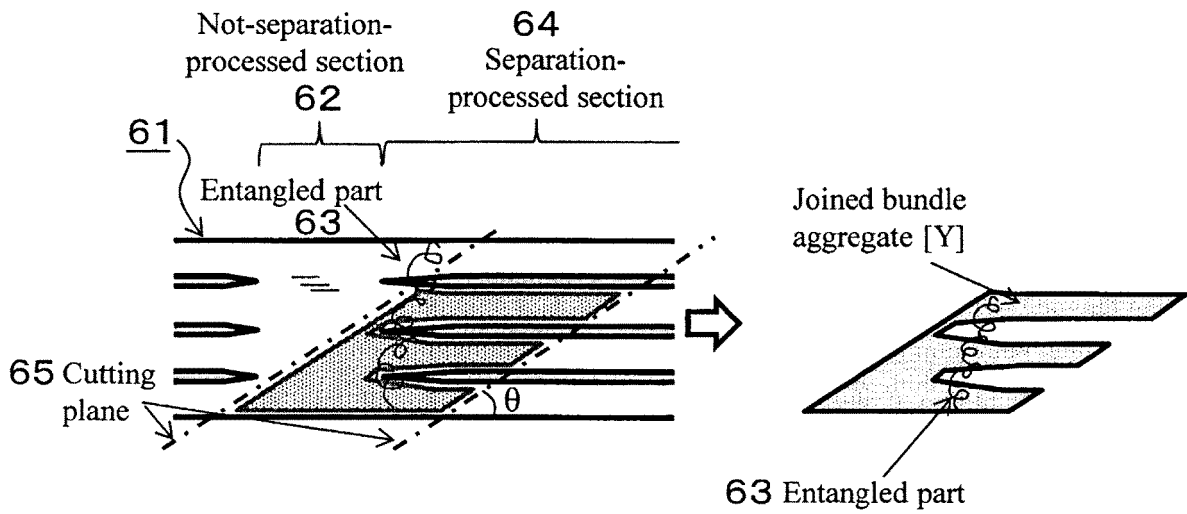


FIG. 11

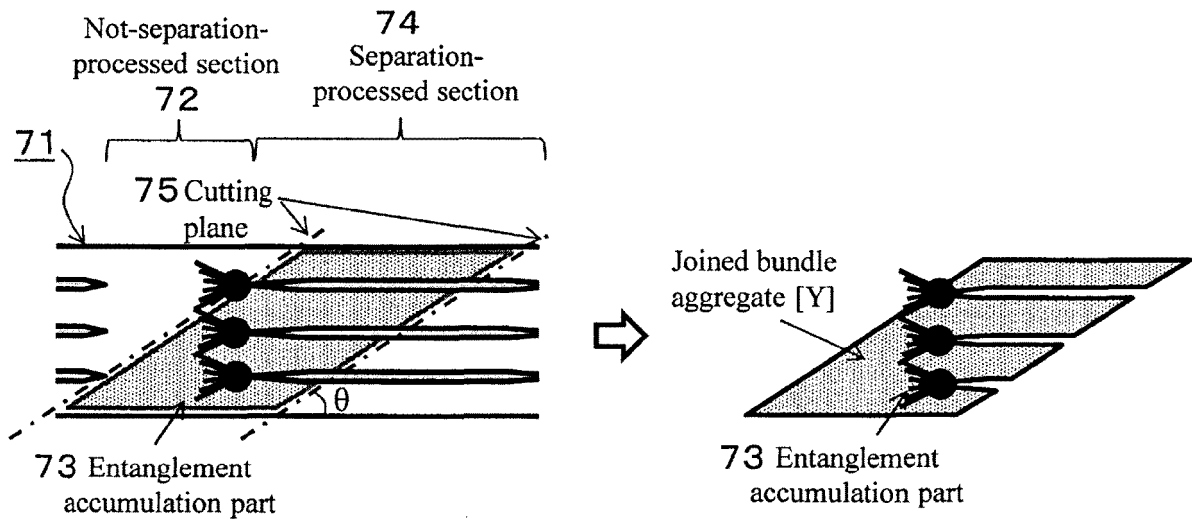


FIG. 12

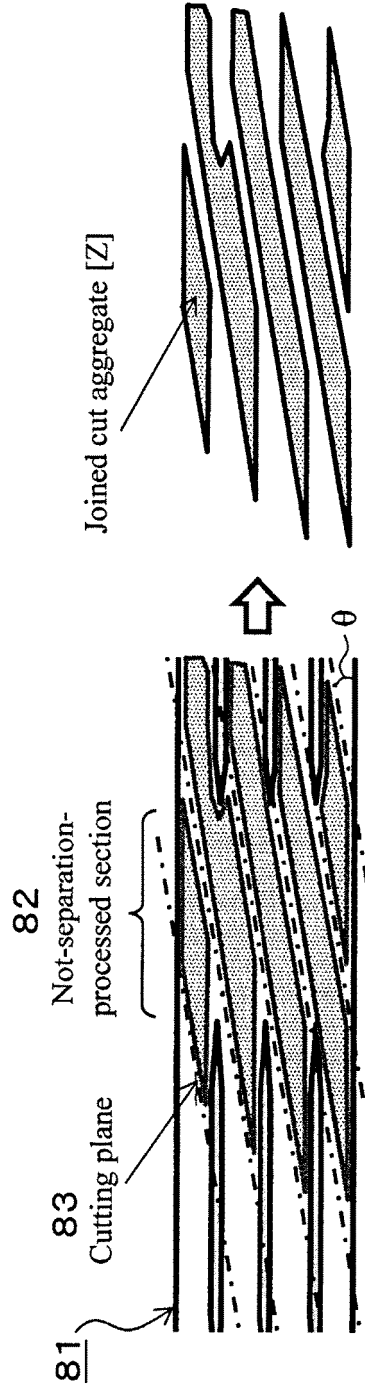


FIG. 13

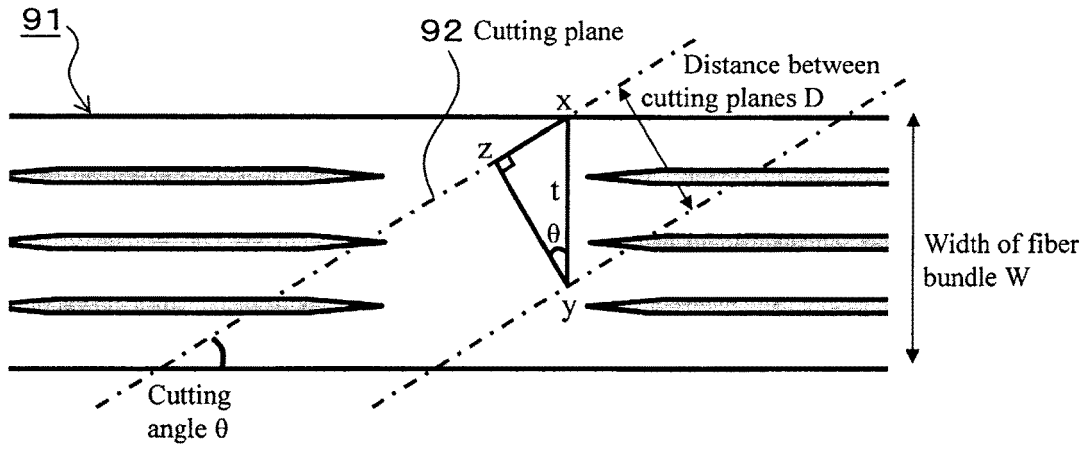
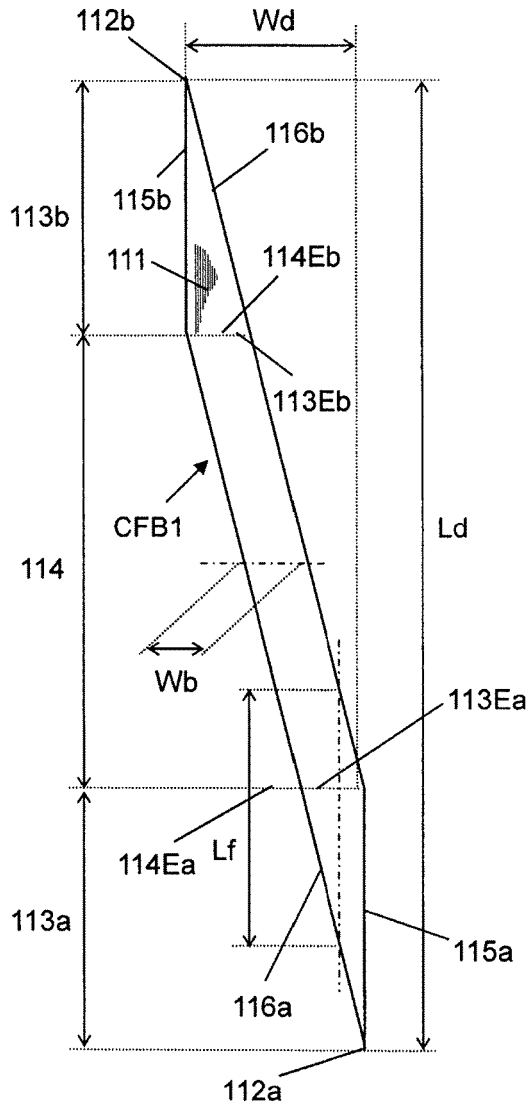
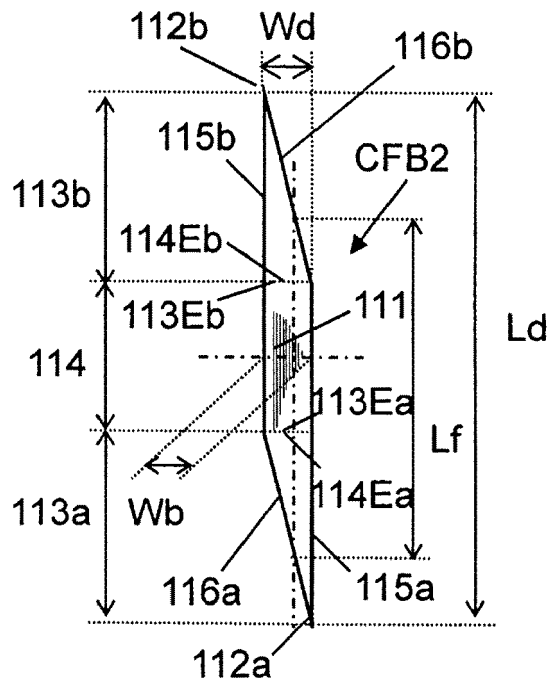


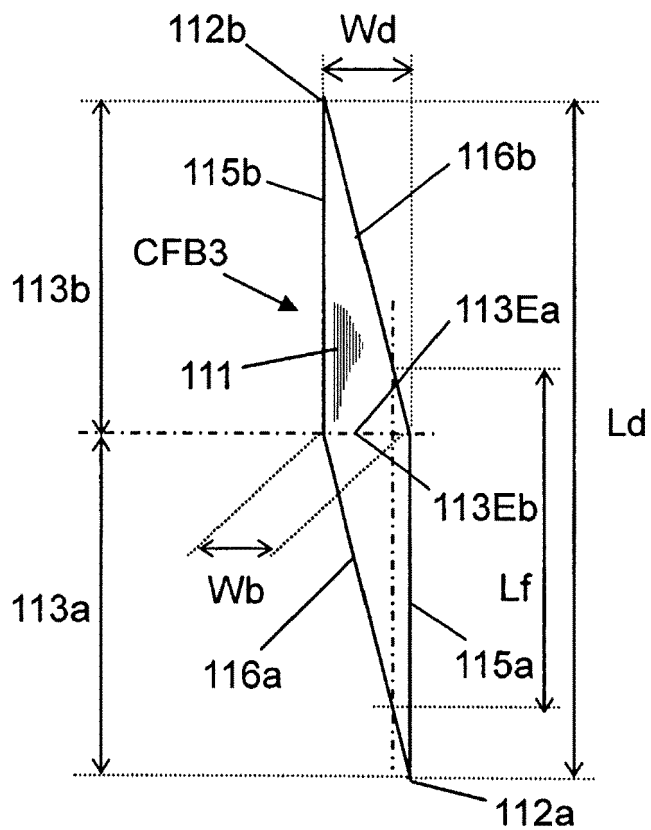
FIG. 14



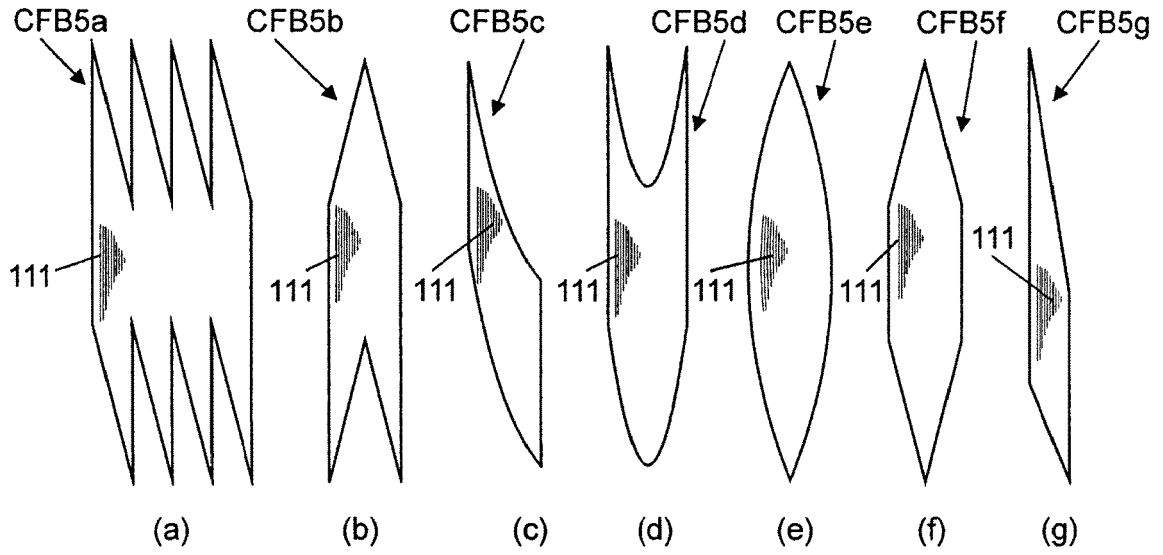
**FIG. 15**



**FIG. 16**



**FIG. 17**



**FIG. 18**

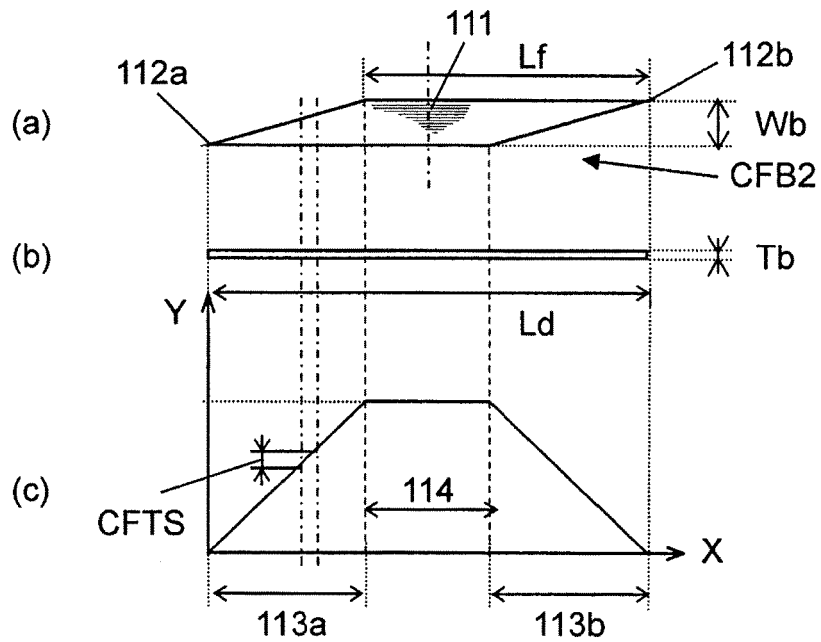


FIG. 19

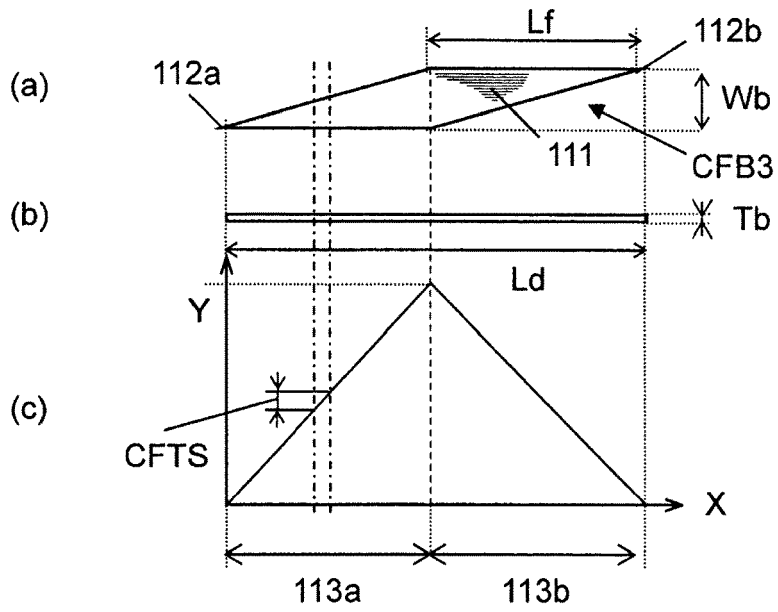


FIG. 20

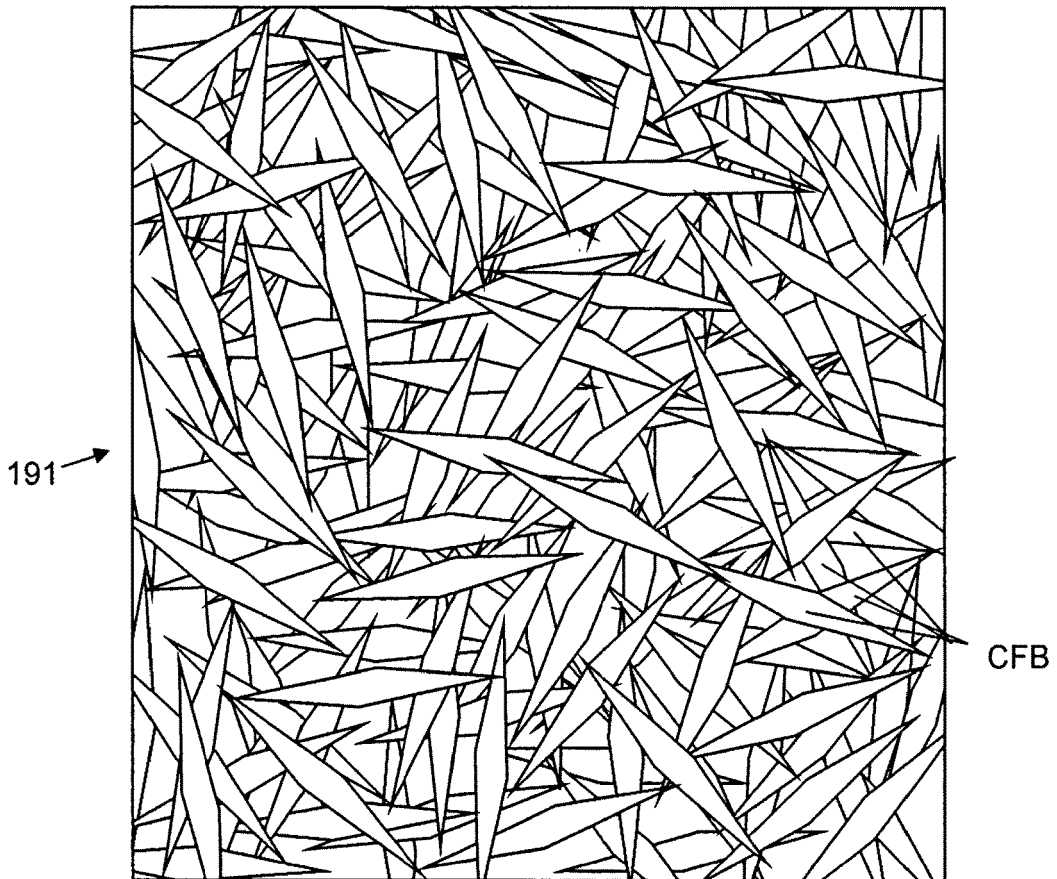
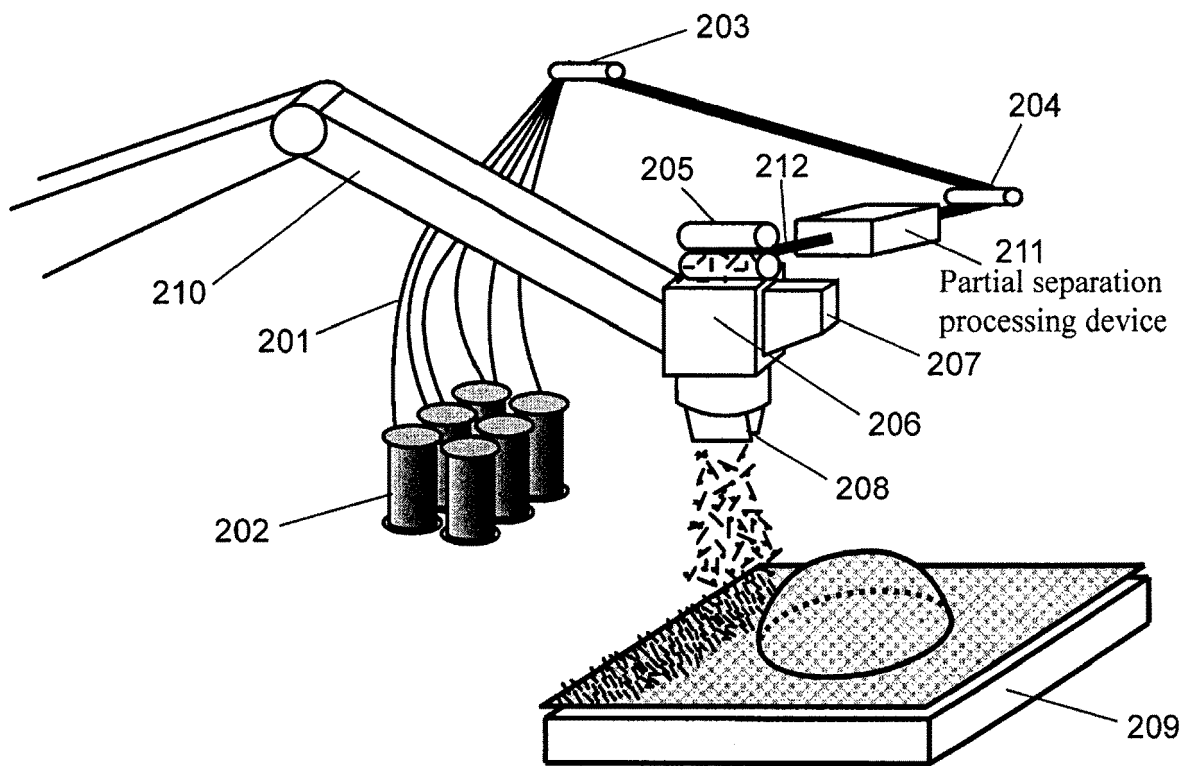


FIG. 21



**REFERENCES CITED IN THE DESCRIPTION**

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