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(54) **Process for producing carbon fiber fabrics.**

(57) A carbon fiber fabric is produced by making carbon fibers having a tensile strength exceeding 250 kgf/mm^2 and an elongation to break of 0.4% to 10% into a two- or three-dimensional fabric, said carbon fibers having been obtained by melt-spinning a carbonaceous pitch, making the resulting fibers infusible and then subjecting the thus-infusiblized fibers to a primary heat treatment at a temperature exceeding 650°C and not higher than 2,500°C, and subjecting said two- or three-dimensional fabric to a secondary heat treatment at a temperature which is not lower than 700°C and is higher than the temperature used in said primary heat treatment.

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Process for Producing Carbon Fiber Fabrics

Background of the Invention

The present invention relates to a process for producing carbon fiber fabrics.

As methods for producing carbon fiber fabrics there are known a method of weaving carbon fibers as a finished product and a method in which an intermediate product is subjected to weaving and the resulting fabric is carbonized or graphitized. As an example of the latter there is disclosed in Japanese Patent Laid-Open No.120136/1988 a three-dimensional fabric containing a pitch based carbon fiber as one component thereof, the carbon fiber having, before heat treatement in a relaxed state, a strength of 15 to 250 kgf/mm^2, an elongation to failure of 0.5% to 8.0% and an elastic modulus of 400 to 40,000 kgf/mm^2, but, after the said heat treatment, capable of increasing in both strength and elastic modulus to 1.1 times as high as the strength and elastic modulus before the heat treatment and capable of a strength of 150 kgf/mm^2 or higher and an elastic modulus 40,000 kgf/mm^2 or higher.

In Japanese Publication No.20281/1987 there is disclosed a process for producing a carbon fiber product in which as-spun pitch fibers are subjected to an initial carbonization treatment, then a bundle of the fibers is subjected to weaving and the resulting fabric is carbonized or graphitized.

Further, in Carbon-Fiber-Reinforced Plastics, Bamberg, West Germany, May 11-12, 1977, there is a description to the effect that infusiblized fibers can be woven if they have a strength of about 40 kgf/mm^2 and elongation to failure of 5%, and can be carbonized into a carbon fiber fabric.

However, in producing fabrics according to the aforementioned conventional methods, breaking of fibers of fuzzing occurs because the strength of the fibers in the weaving stage is not sufficiently high, and therefore even if the resulting fabric is carbonized or graphitized, it is impossible to obtain a high fiber volume fabric. Also a permanent strain may remain after carbonization, so the inherent strength cannot be developed when used as a composite material.

Summary of the Invention

It is the object of the present invention to overcome the above mentioned problems, particularly to provide a process capable of efficiently producing carbon fiber fabrics with little fuzzing and free of permanent strain.

The present invention is concerned with a process for producing a carbon fiber fabric, which process comprises making carbon fibers having a tensile strength exceeding 250 kgf/mm^2 and an elongation to break of 0.4% to 10% into a two- or three-dimensional fabric, the said carbon fibers having been obtained by melt-spinning a carbonaceous pitch, making the resulting fibers infusible and then subjecting and thus-infusiblized fibers to a primary heat treatment at a tempera-ture exceeding 650 °C and not higher than 2,500 °C, and sub-jecting the two- or three-dimensional fabric to a secondary heat treatment at a temperature which is not lower than 700 °C and is higher than the temperature in the primary heat treatment.

Detailed Description of the Invention

The process for producing carbon fiber fabrics according to the present invention will be described in detail hereinunder.

As the carbonaceous pitch there is used a coal or petroleum pitch having a softening point of 100 °C to 400 °C, preferably 150 ° to 350 °C. Both optically isotropic and anisotropic pitches are employable examples of the carbonaceous pitch, but particularly preferred is an optically anisotropic pitch having an optically anisotropic phase content of 60% to 100%.

The melt spinning may be carried out by any suitable known method. The resulting pitch fibers are then rendered infusible.

The infusiblization treatment may be performed at a temperature of 50 °C to 400 °C, preferably 100 ° to 350 °C, in an oxidizing gas atmosphere. As the oxidizing gas there may be used air, oxygen, nitrogen oxide, sulfur oxide, halogen, and mixtures thereof.

The primary heat treatment is conducted in an inert gas at a temperature exceeding 650 °C and not higher than 2,500 °C, preferably in the range of 1,000 ° to 2,000 °C, more preferably 1,500 ° to 2,000 °C.

The treatment time is selected suitably so as to obtain primary heat-treated fibers having later-described tensile strength and elongation to failure, but usually it is in the range of 1 second to 10 hours. According to a method wherein fibers which have been treated at a temperature lower than the above range are subjected to weaving, followed by heat treatment at a high temperature, there remain so-called permanent strain or permanent deformation of the fibers because of a low carbonized state, and thus since the fibers are woven in a bent state, breakage will result at inflection points if pulled.

The primary heat-treated fibers, obtained by going through the above primary heat treatment, have a tensile strength exceeding 250 kgf/mm^2 and a breaking elongation of 0.4% to 10%.

It is an essential condition that the tensile strength of the primary heat-treated fibers should be in excess of 250 kgf/mm^2. Preferably, the said tensible strength is not lower than 285 kgf/mm^2, more preferably not lower than 300 kgf/mm^2, and most preferably not lower than 330 kgf/mm^2. Although there is no upper limit, the tensile strength in question is usually not higher than 1000 kgf/mm^2. If the tensile strength is outside the range just specified, there will occur breakage of fibers and fuzz during weaving, with the result that a high fiber volume fabric cannot be obtained.

The elongation to failure is in the range of 0.4% to 10%, preferably 0.6% to 10%, more preferably 0.6% to 5%. In the case of a three-dimensional fabric it is preferable that the elongation to failure be not lower than 0.6% because of a small radius of curvature of bundle in a fabric.

The value of elastic modulus is determined optionally according to the combination of the above tensile strength and elongation to failure, but usually it is in the range of 5 to 100 x 10^3 kgf/mm^2.

The fiber diameter is in the range of 3 to 100 μm, preferably 5 to 30 μm.

In the present invention, the foregoing primary heat-treated fibers are made into a two- or three-dimensional fabric. Examples of the "fabric" as referred to herein are fabrics obtained using 100 to 25,000 continuous filaments. More concrete examples include two-dimensional fabrics such as plain weave, satin weave, twill weave, bias weave fabrics braid, and stitch knit, three-dimensional fabrics such as three-dimensional orthogonal fabric, leno, interlock and braid, as well as fabrics reinforced in three or more directions such as special shape fabrics, mat-like fabric and felt-like fabric.

The fabric of the primary heat-treated fibers is subjected to a secondary heat treatment. The secondary heat treatment is performed at a temperature which is not lower than 700 °C, for example in the range of 700 °C to 3,300 °C, preferably 1,000 °C to 3,000 °C, more preferably 1,500 °C to 2,800 °C, and which is higher than the temperature in the primary heat treatment. Usually, the secondary heat treatment temperature is higher by 50 °C or more preferably by 100-2,000 °C, more preferably by 200-1,000 °C, than the primary heat treatment temperature. The treatment time in the secondary heat treatment is selected optionally for obtaining the secondary heat-treated fabric falling under the scope of the present invention, but usually it is in the range of 1 second to 10 hours.

According to the process of the present invention there can be obtained a carbon fiber fabric with little fuzzing and free of permanent strain.

<Examples>

The following examples are given to illustrate the present invention more concretely.

Examples 1 - 6

A carbonaceous pitch was melt-spun and the resultant fibers were rendered infusible. The fibers thus infusiblized were subjected to a primary heat treatment at temperatures ranging from 1,700 °C to 2,450 °C. Using the fibers thus heat treated, three-dimensional orthogonal fabrics were produced. Then, the fabrics were each subjected to a secondary heat treatment at 2,500 °C. The three-dimensional fabrics thus heat treated were evaluated, the results of which are as shown in Table 1.

Comparative Examples 1 - 3

Three-dimensional orthogonal fabrics were produced using the primary heat-treated fibers shown in Table 1, and then subjected to a secondary heat treatment at 2,500 °C. The three-dimensional fabrics thus heat treated were evaluated, the results of which are as set forth in Table 1.

Table 1

	Primary Heat-Treated Fibers				Evaluation of three-dimensional fabrics after sencondary heat treatment at 2,500 °C
	Tensile Strength kgf/mm^2	Elongation to failure %	Elastic Modulus 10^3kgf/mm^2	Treating Temp °C	
Ex. 1	280	0.7	40	2100	Good
2	300	0.75	40	2100	Good
3	330	0.83	40	2100	Good
4	370	1.2	30	2000	Good
5	430	1.3	33	2000	Good
6	350	0.7	50	2450	Good
Comp.Ex.1	205	0.7	29.3	1700	Fuzz
2	245	0.5	48.8	2450	fuzz
3	10	3.0	0.3	600	Permanent strain

Claims

1. A process for producing a carbon fiber fabric, which process comprises making carbon fibers having a tensile strength exceeding 250 kgf/mm^2 and an elongation to break of 0.4% to 10% into a two- or three-dimensional fabric, said carbon fibers having been obtained by melt-spinning a carbonaceous pitch, making the resulting fibers infusible and then subjecting the thus-infusibilized fibers to a primary heat treatment at a temperature exceeding 650°C and not higher than 2,500°C, and subjecting said two- or three-dimensional fabric to a secondary heat treatment at a temperature which is not lower than 700°C and is higher than the temperature used in said primary heat treatment.

2. A process as set forth in Claim 1, wherein said carbonaceous pitch is an optically anisotropic pitch having an optically anisotropic phase content of 60% to 100%.

3. A process as set forth in Claim 1 or Claim 2 wherein the fibers after said primary heat treatment have a tensile strength of not lower than 300 kgf/mm^2.

4. A process as set forth in any one of the preceding claims wherein the fibers after said primary heat treatment have an elongation to break in the range of 0.6% to 10%.

5. A process as set forth in any one of the preceding claims wherein the temperature used in said secondary heat treatment is higher by at least 50°C than the temperature used in said primary heat treatment.

6. A process as set forth in any one of the preceding claims wherein the temperature used in said secondary heat treatment is higher by 100° to 2,000°C than the temperature used in said primary heat treatment.