METHOD OF PRODUCING TAPES OF LONGITUDINALLY ALIGNED CARBON FIBRES

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10 Claims

ABSTRACT OF THE DISCLOSURE

A method of producing tapes of carbon fibres in which a sheet of longitudinally aligned polyacrylonitrile fibres is pyrolysed and subsequently divided longitudinally to form a plurality of tapes of the carbon fibres thus produced.

This invention relates to a method of producing tapes of longitudinally aligned carbon fibres.

In the various winding processes for the production of artefacts from fibre reinforced materials, it is often found that the most convenient form for the fibrous material to be wound comprises a tape of longitudinally aligned fibres. Over recent years, carbon fibres have been introduced as fibrous reinforcements, and there has grown a demand for such fibres in tape form.

The present invention provides a method of manufacturing such a tape of carbon fibres.

According to the present invention, a method of producing tapes of longitudinally aligned carbon fibres comprises the pyrolysis of a sheet of longitudinally aligned polyacrylonitrile fibres and the subsequent division of the sheet into a plurality of tapes.

Preferably said pyrolysis is preceded by a pre-oxidation step in which the fibres are heated in air.

The pyrolysis is preferably followed by a resin impregnation step in which the sheet of fibre is impregnated with a resin matrix material, and said division preferably takes place after said impregnation.

The pre-oxidation heat treatment may take place with the fibre heated to between 150° C. and 300° C.

Said division may be effected by a plurality of knifedge slitting wheels; thus the wheels may comprise two or more separate sets which effect the desired splitting in two or more stages.

The invention will now be particularly described merely by way of example with reference to the accompanying drawings in which FIG. 1 is a flow diagram of apparatus for carrying out the method of the present invention, and FIG. 2 is a perspective view of the sheet slitting apparatus of FIG. 1.

In FIG. 1, 10 indicates a creel unit in which a plurality of creels 11 hold tows of polyacrylonitrile fibres of 1½ denier. In this creel unit there is embodied a sheet forming and retaining unit (not shown) such for instance as is described in co-pending application Ser. No. 759,730, filed Sept. 13, 1968, now abandoned. The creel unit produces sheets held together by a weft thread introduced at intervals in the warp fibres, each sheet being of approximately 18 inches width and each containing 50 tows, each tow comprising 10 fibres. The eight sheets are maintained with a separation of some three inches between sheets, the sheets lying one above the other, and are fed into a roller unit generally indicated at 12. The roller drive unit 12 comprises eighteen separate sets of four rollers, one of which is indicated at 13. These sets of rollers are mounted in parallel so that each roller set receives a single sheet which then passes round the four driven rollers which comprise the set and then passes out from the drive unit horizontally aligned with its entry.

On passing from the drive unit 12 the eight sheets are brought closer together as shown at 14 and pass through closely fitting orifices at 15 into a pre-oxidation furnace 16. The atmosphere inside the furnace is maintained by a blower system and heater units (not shown) so that the ambient atmosphere within the furnace comprises air heated to a temperature of some 220° C. A blower system 17 is provided to effect a controlled flow of air in the furnace.

Within the furnace 16 are a plurality of sets of rollers indicated by 18. The eight sheets pass over these rollers which cause them to move over a castellated path within the furnace, in this way achieving a long path length (40 feet) within a relatively compact furnace; the sheets are arranged to take some seven hours to pass through the furnace.

While within the furnace 16 the fibres are therefore pre-oxidised, that is some degree of modification takes place between individual molecules to stabilise their structure.

On emerging from the furnace through closely fitting orifices at 19 the fibres, still in their sheet form, pass into a second roller drive unit 20, which is identical to that shown at 12. By arranging the drive units 12 and 20 so that either end of the furnace 16 the fibre can be arranged to be held under tension while it is being pre-oxidised. This prevents the shrinkage which would otherwise take place during this stage of the process.

On leaving the drive unit 20 the sheets are brought together to form a single bunch and pass into a carbonising furnace 21 by way of a roller seal unit 22. This seal unit makes use of a pair of roller seals as described in co-pending application Ser. No. 771,466, filed Oct. 29, 1968, now abandoned and prevents any considerable leakage from or into the furnace 21. The fibre sheets are compacted at this stage to reduce the power required to simplify the system; at this stage it is unnecessary to maintain the spacing which is required in the previous stages to allow purging of reaction products and dissipation of heat.

In the furnace 21 a nitrogen atmosphere is maintained, a duct 23 being provided for the influx of nitrogen while a duct 24 is provided for the efflux of nitrogen plus waste gases. Inside the furnace 21 the sheets are suspended above a floor (not shown). Within the furnace the temperature is arranged to rise steadily from room temperature adjacent to seal unit 22 up to a furnace temperature of 300° C. at a point 25. The dimensions of the furnace are so chosen that at the point 25 the majority of the pyrolysis of the fibres has taken place; this being so there is no necessity to arrange for any additional purging of reaction products in the subsequent treatment.

The compacted fibres next pass through a higher temperature portion 26 of the furnace. It will be noted that the sectional area of this part of the furnace is reduced by wall assemblies 27 which comprise heating units; this reduction in area helps to reduce the power required to provide the higher temperature heating. In the section 26 the fibres are further heated to a temperature in the region of 1600° C.

The compacted fibres finally emerge from the furnace 21 by way of a second seal unit 28 which again uses a roller seal.

On leaving the seal unit 28 the fibres pass into a further furnace 29. Although for convenience of drawing, this furnace is shown as being separate from the furnace 21, it is in fact connected directly to the seal 28 and the entire apparatus forms a linear layout. Thus the seal 28 in fact divides the atmosphere within the furnace 21 from that in the furnace 29. Within the furnace 29 an atmosphere of argon is maintained and once again an input
3 pipe 30 supplies argon while an output pipe 31 allows egress of argon and any products of reaction. Within the furnace 29 the fibre is heated, still in its compacted form, to a temperature in this case of 2500° C, but which may be in the range of 1500° C. upwards. In this stage the structure of the carbon fibre is not changed, only its extent. Graphic-base this stage can in fact be omitted completely and it is possible to produce useful carbon fibre with a carbonising furnace which merely heats the fibre up to a temperature of 1000° C to 1200° C.

Then from the furnace 29 after some 1½ hour residence time by way of a seal unit 32 which is again similar to the unit 28 and prevents large escape of gases from the furnace 29. Immediately on leaving the seal unit 32 the compacted fibres pass between a pair of driven rollers 33 and after passing through these rollers the compacted fibres are again split up into their eight discrete sheets into a resin impregnation unit generally indicated at 34. The resin impregnation unit 34 is of the type described and claimed in co-pending application Ser. No. 887,864, filed December 24, 1968, now abandoned and uses coated paper as a transferring agent for an epoxy nolacite resin. For simplicity, the course of a single sheet will be described; the remaining sheets follow an exactly similar course.

Considering the upper sheet first, on entering the unit 34 it passes between a pair of reels 35 and 36 of resin coated release paper. The release paper is itself impregnated with a release agent which allows the resin to be transferred to the paper. In order to make the coated paper easier to handle, the reels 35 and 36 are mounted within a chamber 37 which is refrigerated so that the resin is maintained non-tacky.

The fibre from the reels 35 and 36 is fed on to either side of the fibre sheet so that the coated side of the paper is against the fibre. The “sandwich” thus formed passes from the chamber 37 into a second chamber 38 and between a pair of heated rollers 39. On passing between these rollers, the combined effect of the heat and pressure evolved causes the resin to transfer from the release paper to the fibre sheet, thus impregnating the sheet with resin.

Each sheet then passes through a separate slitting device; only the device for the sheet passing through the rollers 40 is shown on the drawing, and is generally indicated at 41.

The slitting device 41 comprises two pairs of rollers 42 and 43, first and second sets of slitting wheels 44 and 45, which are rotated by drive means (not shown), and a take-up roller 46. The sheet passes between the pairs of rollers 42 and 43, which maintains its overall sheet form, and intermediate these rollers the first set of slitting wheels 44 is mounted so that the saw-toothed wheels divide the sheet, in the present instance into three equal longitudinal portions.

The sheet thus divided passes through the rollers 43 and is then further divided by the slitting wheels 45. In the present case the wheels 45 are mounted to divide the sheet (already divided into three parts) into six tapes.

To prevent clogging of the wheels by resin used to impregnate the fibre, cold air from a vortex tube is arranged to circulate in the vicinity of all the slitting wheels.

The sheet is thus formed into six tapes, and these tapes are then rolled on the take-up roller 46 to form six reels of tape on a single roller. The roller may then be used to store the tapes, or may be itself divided into six parts carrying one tape reel.

The reel of tape thus produced may be used in various ways, particularly for the laying-up of composite articles by winding techniques.

We believe that the size roll on the unimpregnated stage 41 may be used at any stage of the carbon fibre forming process described above but subsequent to the carbonising stage which takes place in the furnace 21. If the device 41 is interpolated at any of the suitable positions “upstream” of the impregnation unit 34, it becomes necessary to maintain the separation of the tapes throughout the following treatment stages which could be difficult.

To produce unimpregnated tapes dry fibre is collected on a reel just prior to the impregnation stage and subsequently split by stationary knives in device 41 and wound onto smaller reels more which it may subsequently be reprocessed through 41 to produce still narrower tapes e.g. 10,000 filaments. Such stationary knives are preferably angled to the longitudinal axis of the sheet in such a way that any snap not immediately cut by the knife causes the sheet to ride up the knife and cause the effect of gravity to add to the cutting effect.

Again the device 41 is only one way of dividing the sheet into tapes; various forms of stationary or moving knives or other dividers could be used, and the take-up arrangement could for instance comprise one or more rollers or further transverse slitting devices which produce stacks of the tapes cut to a predetermined length.

It is quite possible to use other resins than epoxides, and it may be that with some resins it will be unnecessary to cool the cutters, which may be of saw-toothed.

Although the embodiment described has the slitting machine 41 as an integral part of the production line, it would of course be possible to have the device 41 “off-line.” In this case, on leaving the rollers 39, each sheet is wound onto a reel the lower release paper being pulled away so as to wind only fibre plus top release paper. The fibre is wound so as to be under the release paper.

The reel of fibre plus release paper is then taken to a separate slitting machine 41.

At this stage it may be convenient to replace the release paper by another paper or plastic film, which may be easily extensible and compressible, e.g. may be embossed or crinkled. This extensibility helps winding, while the compressibility helps to compensate for variations in sheet thickness. The re-winding is preferably performed with an edge guide so as to produce an accurately square-edged roll and make the slitting process more accurate.

We claim:
1. A method of producing a tape of longitudinally aligned carbon fibres comprising the steps of:
   (a) providing a sheet of a plurality of longitudinally aligned polycapronitrile fibres;
   (b) pyrolyzing said sheet of step (a) in an inert pyrolyzing atmosphere and at elevated temperature and thereby converting said polycapronitrile fibres to carbon fibres;
   (c) impregnating the thus pyrolyzed sheet with a resin material; and thereafter
   (d) dividing the resin-impregnated pyrolyzed sheet into a plurality of tapes;
2. A method as claimed in claim 1 and in which said pyrolysis is preceded by a pre-oxidation step in which the polycapronitrile fibres of step (a) are heated in air to a temperature between 150° C. and 300° C.
3. A method as claimed in claim 1 and in which said impregnation is performed by transfer of resin from resin coated release paper to the sheet of fibres.
4. A method as claimed in claim 1 and in which said division takes place after said impregnation, the release paper being divided along with the fibre sheet.
5. A method as claimed in claim 1 and in which said division is effected by a plurality of slitting wheels.
6. A method as claimed in claim 5 and in which there are two sets of wheels, one set being spaced apart from the other set, which divide the sheet into successively smaller tapes.
7. A method as claimed in claim 1 and in which said pyrolysis takes place in a continuous process, the fibre sheet being wound up after said pyrolysis and removed to a separate slitting machine.
8. A method as claimed in claim 7 and in which said wound fibre sheets are interleaved with a form of release paper.
9. A method as claimed in claim 1 in which an epoxy novolac resin is applied in step (c).

10. A method of producing a tape of longitudinally aligned carbon fibers comprising the successive steps of:

(a) providing a sheet of longitudinally aligned polyacrylonitrile fibers by assembling a plurality of tows comprising individual polyacrylonitrile fibers into a sheet of material;

(b) preoxidizing the fiber sheet of step (a) in heated air at a temperature of about 150° C. to about 300° C.;

(c) pyrolyzing the preoxidized sheet of step (b) in an inert atmosphere at a temperature of at least 1000° C. and thereby converting the polyacrylonitrile fibers to carbon fibers;

(d) impregnating the thus pyrolyzed sheet of fibers of steps (c) with a resin material by transferring the resin material from a resin coated release paper to the pyrolyzed sheet of fibers; and thereafter

(e) dividing the resin-impregnated pyrolyzed sheet into a plurality of tapes.