March 19, 1974 J. N. HALL 3,798,095

PROCESS FOR SPREADING A GRAPHITE FIBER TOW INTO A RIBBON OF GRAPHITE FILAMENTS

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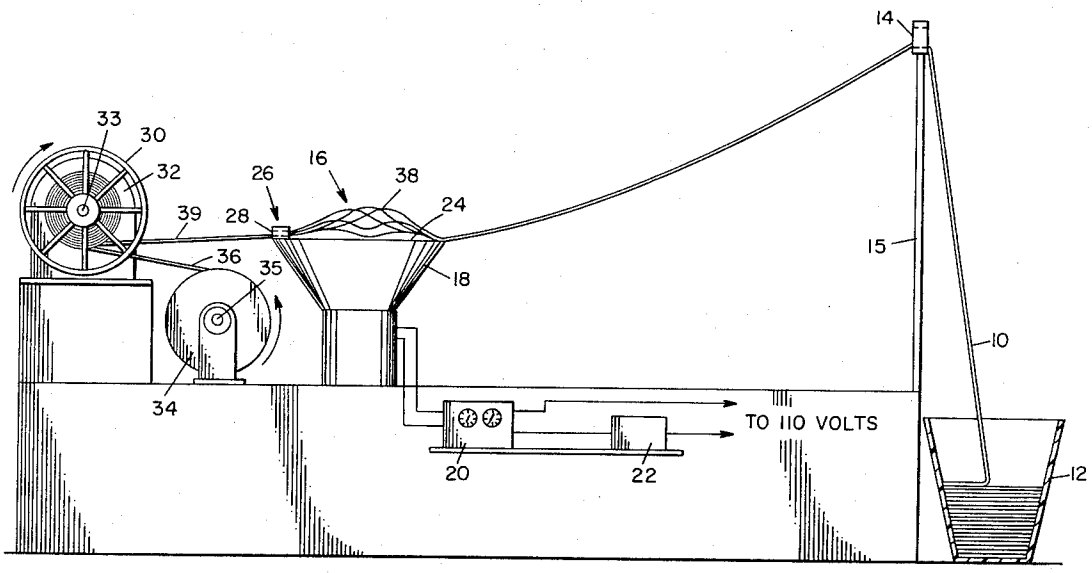


FIG. 1

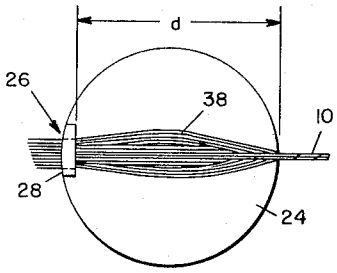


FIG. 2

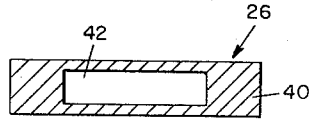


FIG. 4

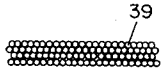


FIG. 3

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PROCESS FOR SPREADING A GRAPHITE FIBER TOW INTO A RIBBON OF GRAPHITE FILAMENTS

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Int. Cl. B32b *31/16*

U.S. Cl. 156—73 5 Claims

ABSTRACT OF THE DISCLOSURE

A process for spreading a graphite fiber tow into a mass of substantially individual graphite filaments is provided. Spreading of the filaments is achieved by vibrating the graphite tow in air pulsating at a frequency and intensity sufficient to couple the energy of the pulsating medium to the graphite tow. Vibration of the tow in air causes separation and spreading of the filaments in the tow.

This is a division of application Ser. No. 97,988, filed Dec. 14, 1970, now U.S. Pat. No. 3,704,485.

This invention relates to a process for spreading a graphite fiber tow into a ribbon of graphite filaments. In another aspect, this invention relates to a process for preparation of a graphite fiber tape from a graphite fiber tow. In still another aspect this invention relates to a system for spreading a graphite fiber tow into a ribbon of graphite filaments having a controllable width and filament density.

A tow is a loose rope of textile filaments. The filaments in a graphite fiber tow each have a diameter of between about 8 to about 10 microns. A graphite fiber tow generally contains from about 8,000 to about 10,000 individual filaments in the tow. Graphite fibers are anisotropic materials, i.e., they are fibers exhibiting different property values when the properties are evaluated along different axes of the material.

For many applications in which graphite fibers are to be employed, it is desirable that the graphite fibers be available in the form of a thin ribbon of filaments in which substantial numbers of the filaments are in the same plane and in which the filaments are in a parallel and side-by-side relationship. Thus, it is desirable to be able to convert a graphite fiber tow into a ribbon of filaments in which the filament density of the ribbon is controllable and the width of the ribbon is uniform. A ribbon of graphite filaments, as heretofore described, can be readily impregnated with a resinous material which man be solidified through curing or cooling depending on the resin type to provide a unitary and solid ribbon of graphite filaments which can be marketed commercially in the form of a graphite tape.

A process has now been discovered for spreading of a graphite fiber tow into a ribbon of filaments in which filament density and width is reproducible and controllable. This ribbon of filaments can be employed for manufacture of broad goods or it can be contacted directly with a resin to produce a self-supporting tape of the desired width and filament density. The process of this invention comprises drawing a graphite fiber tow under low tension through a pulsating gaseous medium, said pulsations being generated in said gaseous medium at a frequency and intensity sufficient to couple the energy from the pulsating gaseous medium to the graphite fiber tow whereby the graphite filaments in the tow are made to vibrate with a frequency substantially equal to that of the pulsating gaseous medium spreading the tow into a ribbon of substantially individual graphite filaments. In preparing a

tape, the spread graphite filaments are formed into a ribbon of graphite filaments having a substantially constant width and depth and the ribbon is contacted with a resin capable of bonding the filaments together. The resin-wet filaments are then bonded together and a self-supporting tape of filaments having a substantially uniform depth or filament density and ribbon width is recovered. The process of this invention is more fully described with reference to a process for making a tape of graphite fibers in the following drawings. In the drawings like numbers refer to like parts where applicable.

FIG. 1 is a schematic diagram illustrating typical equipment employed in conducting the process of this invention.

FIG. 2 is a schematic top view illustrating the appearance of a graphite tow as it is being spread by a pulsating column of gas generated by a vibrating surface as hereinafter more completely described.

FIG. 3 is a schematic end view illustrating the appearance of a typical ribbon of graphite filaments prepared in accordance with the process of this invention.

FIG. 4 is a cross-sectional view of a guide means for controlling the width of the spread tow.

In FIG. 1 a graphite fiber tow **10** is illustrated being drawn from a storage container **12** through a centering ring **14** mounted at the top of support member **15**. The centering ring **14** is positioned above the pulsating gaseous medium **16** through which the graphite fiber tow **10** is drawn. The size of the pulsating gaseous medium **16**, referred to sometimes hereinafter as an air mass, through which the graphite fiber tow is drawn is determined by the size and configuration of the vibrating surface employed in conjunction with a vibrator means for inducing pulsations into the air mass. As illustrated in FIG. 1, the vibrator means employed is a loud speaker **18** which is connected through an amplifier **20** to an audio signal generator **22**. A speaker covering is stretched tightly across the speaker cone forming the surface **24**. A guide means **26** is secured at one side of the speaker, referred to herein, as the output-end **28** of the speaker **18**. The graphite fiber tow **10** is secured to a wind-up spool **30** which is driven by a conventional drive means **32**. The wind-up spool **30** is driven about its longitudinal axis **33** by drive means **32** in the direction shown by the arrow. A second spool **34** is mounted for rotation about its longitudinal axis **35** and contains paper **36** coated with resin. The second spool is mounted between the output-end **28** of the speaker **18** and the wind-up spool **30**. Second spool **34** releases resin coated paper in response to pulling force of the wind-up spool **30**. Paper **36** from the second spool **34** is secured to the wind-up spool **30** in alignment with the graphite tow **10** which is to be separated into a ribbon of filaments.

FIG. 2 illustrates the graphite fiber tow **10** being spread into a mass of individual filaments **38** as the tow **10** passes through the pulsating air mass directly over vibrating surface **24**. The linear distance through the pulsating air mass through which the graphite fiber tow **10** is drawn is referred to as the free span of the fibers. The spread tow **38** passes through guide means **26** at the output-end **28** of the speaker **18** wherein the mass of individual filaments **38** is formed into a ribbon **39**. The ribbon configuration of the filaments as the filaments pass through guide means **26** forming a ribbon **39** is schematically illustrated in FIG. 3. This drawing (FIG. 3) is intended to be schematic only, since a real tow passing through guide means **26** contains between 8,000 and 10,000 filaments.

A cross-sectional view of guide means **26** is shown in FIG. 4. Guide means **26** consist of a guide means body **40** having a conduit **42** therethrough. The width of conduit **42** through guide means body **40** establishes the width of the ribbon of spread graphite filaments produced in accordance with the process of this invention.

Description of the operation of the graphite filament spreading

To initiate the spreading process of this invention the signal generator oscillator **22** and amplifier **20** are energized by closing the circuit connecting them to a power source such as 110V A.C. source. Signal generator oscillator **22** generates a signal having the desired frequency. Amplifier **20** receives the signal from the signal oscillator generator **22** and increases the intensity of the signal to a suitable level. Speaker **18** is connected to amplifier **20**. The speaker **18** receives the amplified signals in the form of electrical impulses and transforms these impulses into acoustical energy waves without a substantial change in the wave form. Acoustical energy radiates from the speaker **18** in the form of sound waves, which establish vibrations in the surface covering **24** of the speaker **18** and in the air mass directly above the speaker. The pulsating speaker surface **24** functions as a pulstating pump to cause the air mass directly above the speaker to pulsate. After the air mass over the speaker has started pulsating, the wind-up spool **30**, to which has been secured both the free-end of a graphite fiber tow **10** and the free-end of the resin impregnated paper **36**, is actuated. Graphite fiber tow **10** is drawn through the pulsating air mass directly above vibrating surface **24** of speaker **18**. The graphite fiber tow **10** being drawn across surface **24** of the speaker **18** vibrates rapidly as it passes through the pulsating air mass. The vibrations in the tow **10** approach the frequency of vibration of the speaker **18** until the multiplicity of the filaments comprising the graphite fiber tow **10** each visually appears to be vibrating separately in a wide dispersion of filaments (spread tow **38**) as schematically shown in FIGS. 1 and 2. The spread tow **38** is drawn by the wind-up spool **30** through a conduit **42** in guide means **26**. The spread filaments are drawn through conduit **42** in guide means **26** forming a ribbon. The width and depth of the filaments comprising the ribbon formed is controlled by the width of the conduit **32** in the guide.

Wind-up spool **30** functions to pull graphite tow **10** through the pulsating air mass and to bring the ribbon of separated graphite filaments into contact with resin coated paper **36**. The wind-up spool **30** is turned by variable speed motor **32** such that the withdrawal rate of graphite tow from the storage vessel is at a linear rate of from about 5–130 ft./min. While both lower and higher rates of withdrawal of the graphite fiber through the pulsating air column can be employed, it is necessary to control the feed rate of the graphite tow through the pulsating column such that the tension within the graphite fiber is low, on the order of about 6 grams or less. Low tension in the graphite fiber tow is necessary when separating the tow into a ribbon of filaments in accordance with the process of this invention. High tension in the tow is undesirable because it makes removal of false twist, oftentimes present in graphite fiber tow, difficult or impossible to achieve. Thus, when conducting the spreading process of this invention, low tension in the graphite fiber tow can be achieved, for example, by passing the tow through a centering ring spaced apart from the vibrator means and above the plane in which the graphite fibers are vibrated in the pulsating air mass. In this manner, the graphite fiber tow being pulled through the pulsating air mass initially comes in contact with the pulsating air mass at an angle which is substantially tangent to horizontal plane through the pulsating air mass. Spacing this centering means apart from and in a plane above the vibrating means, permits the graphite tow to fall to the surface of the vibrating means with substantially no tension on the filaments comprising the tow, with the exception of the tension imposed by weight of each filament. With low tension in the filaments, substantially all of the false twist which occurs naturally in the graphite fiber tow during manufacture will fall out prior to and during spreading of the tow.

The frequency and intensity of the pulsating gaseous medium required to couple the energy of the gaseous medium to the fibers whereby the fibers vibrate in the gaseous medium and are separated thereby, are dependent upon several principal factors. These factors are (1) the tension in the fibers, (2) the free span of the fibers and (3) the characteristics of the speaker which established the pulsations in the gaseous medium. For any given system, the frequency of the pulsations will vary depending on the tension in the fibers. As the tension of the fibers increases, the frequency of the vibrations required to separate the graphite tow will increase. The free span of the fibers is the distance between points at extreme opposite ends of the pulsating medium which are support areas for the fibers and through which the fibrous tow passes into and out of contact, respectively, with the pulsating gaseous medium. As the free span through which the fibers are vibrated increases, the frequency of vibration required to separate the fibers decreases. Another factor to be considered in operating the process of this invention is to evaluate the characteristics of the speaker employed as a vibrating means. The above factors can be readily balanced with a minimum of experimentation following the process of this invention as described herein to arrive at a suitable operational combination for spreading of a graphite fiber tow into a ribbon of filaments.

EXAMPLE

A graphite fiber tow prepared from a polyacrylonitrile precursor fiber is spread into a ribbon of filaments employing the process equipment illustrated schematically in FIG. 1. The equipment employed is as follows.

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Audio signal generator -----	Hewlett-Packard Model 200 CD.
Audio amplifier ----------	50 watt.
Speaker ----------	Electro Voice Model SP15, 80 watt, 4 pounds, 10 ounce magnet, 25–15,000 c.p.s.
Speaker surface ----------	1 mil Mylar film.

The audio signal generator is operated at 108 cycles/second frequency. The guide means employed has a conduit width of 1.0 inch. A centering ring is spaced 15 inches above the speaker surface. The support member on which the centering ring is mounted is spaced 56 inches from input end of the speaker. A graphite fiber tow containing between about 8–10,000 filaments is passed through a system substantially as described. Graphite fiber tow is drawn across the speaker surface at a rate of 20 ft./min. The fibers in the tow are separated into a ribbon of filaments in which most of the filaments are in a parallel and side-by-side relationship.

The system heretofore described for separation of a graphite fiber tow into a ribbon of filaments employs a surface across which the fibers are drawn. The system of this invention is, however, not so limited. For example, the graphite tow need only be drawn over two areas of support for the fibers defining a free span therebetween. Thus, the fibers could be drawn across a speaker not having a surface covering. The opposite sides of the speaker would provide the areas of support necessary for operation of the process of this invention. The gaseous medium through which the fibers are drawn comprises the gaseous environment in which the system is operated which medium would extend below the fibers into the speaker cone and above the fibers. The gaseous medium in which the system is operated is usually air, but it is not limited thereto.

The process of this invention is applicable to spreading of graphite fiber tow prepared from any precursors employed in preparation of graphite fibers such as, for

example, polyacrylonitrile, polyvinyl alcohol, cellulose, regenerated cellulose and the like.

The spread graphite filaments can be bonded together in form of a tape employing any of the well known resins which can be cured or molded under heat and pressure. Exemplary resins which can be employed include epoxy novolaks, polyimides, and epoxies of the two well known types, viz., the bis-phenol epichlorohydrin and the cycloaliphatic types, polyesters, polyurethanes and thermoplastics.

Resins such as those heretofore described, can be applied to the graphite fibers by any suitable method. A suitable method is to provide a film of the resin on a release type paper and to wind the ribbon of graphite fibers onto a wind-up spool in contact with the resin-wet side of the release paper as shown schematically in FIG. 1. If desired, the ribbon graphite fibers can be wet with a solvent for the resin paper prior to contacting the filaments with the resin. Any material which is a solvent for the resin material which is to form the composite with the graphite fibers can be employed. The resin-wet filaments are bonded together into a self-supporting graphite fiber tape by partially curing the resin. The tape thus prepared can be readily stored and handled in this condition. When employed in its final end use, the resin can be completely cured employing heat and pressure. Solvents which can be employed with the exemplary thermosetting resin materials described herein include methylethyl ketone, acetone, ethanol, toluene, and mixtures thereof. It is to be understood that the foregoing methods of applying resin to the spread filaments in preparing a tape and bonding the filaments together are illustrative only, and are not included herein by way of limitation.

Graphite ribbon can be prepared in accordance with this invention in any suitable width and, in particular, in widths varying from about ¾ inch to up to about 2 inches. The width of the ribbon which is prepared from a graphite tow following the process of this invention is controlled by the guide means heretofore described. After the separated filaments pass through the guide means, the filaments are no longer in contact with the pulsating column of air and each filament retains its relative position with the other filaments of the graphite tow in a substantially parallel and side-by-side relationship until these filaments are either wet with resin, wound about the take-up spool and partially cured between resin impregnated release paper, or the ribbon is wound directly around a rotating mandrel to prepare graphite broad goods.

The filament density in the ribbon of graphite fibers produced in accordance with this invention is directly dependent upon the number of filaments in the tow and upon the width of the ribbon produced. Thus, filament density of a graphite fiber ribbon produced from a graphite fiber tow in accordance with the process of this invention is substantially greater when the width of the ribbon produced is narrow. If a graphite tape having a large width of say, for example, about 3 inches and having a high filament density is desired, it is possible to bond a number of graphite tapes having the same width together to form a unitary tape having the requisite filament density.

What I claim and desire to protect by Letters Patent is:

1. A process for preparing a tape of graphite filaments from a graphite fiber tow comprising,

(a) drawing a graphite fiber tow under low tension through a gaseous medium pulsating at a frequency and intensity sufficient to couple the energy from the pulsating gaseous medium to the graphite fiber tow whereby the filaments in the tow are vibrated at substantially the same frequency as that of the pulsating gaseous medium, said pulsation of the fiber spreading the tow into a ribbon of graphite filaments,

(b) contacting the ribbon of filaments with a resin capable of bonding the filaments together and

(c) bonding the resin-wet filaments together, forming a self-supporting tape of graphite fiber.

2. The process of claim **1** in which the ribbon of filaments of step (a) is drawn through a guide means to prepare a ribbon having a substantially uniform width and depth prior to contact of the ribbon with resin.

3. The process of claim **2** in which the graphite fiber tow is prepared from polyacrylonitrile precursor, said graphite fiber tow comprising from about 8,000 to about 10,000 filaments of graphite per tow and in which the graphite filament diameter varies from about 8μ to about 10μ.

4. The process of claim **2** in which the resin employed is capable of bonding the filaments together when partially cured, and the resin is partially cured forming a self-supporting tape of graphite fiber.

5. The process of claim **4** in which the graphite fiber tow is prepared from polyacrylonitrile precursor, said graphite fiber tow comprising from about 8,000 to about 10,000 filaments of graphite per tow and in which the graphite filament diameter varies from about 8μ to about 10μ.

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19—65 T; 117—Dig 8; 156—180