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METHOD OF FABRICATING FIBER-REINFORCED ARTICLES

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ABSTRACT OF THE DISCLOSURE

Filaments formed of silicon carbide-coated boron with an overcoat of a matrix material including such metals as aluminum, magnesium or titanium are formed into an article of appropriate shape and hot pressed to thereby provide a fiber-reinforced article having a high volume percentage of reinforcing fiber, the process being characterized by a high degree of reproducibility.

BACKGROUND OF THE INVENTION

It is known that fiber strengthening offers the potential of significant improvements in the fabrication of structural materials designed to meet the imposing requirements of space age hardware. The concept of fiber strengthening is based on the fact that materials produced in the fibrous form frequently exhibit a larger elastic strain capacity and, hence, higher strength than the corresponding materials in bulk. In order to exploit these properties, it is necessary to gether these fibers together in the desired structure in such a way that failure in several isolated fibers will not be transmitted to the surrounding fibers, and further, to distribute the load with reasonable uniformity over the entire fiber bundle. One method of effecting this result is to encase the fibers in a matrix material which will deform plastically.

One of the paramount problems in obtaining high strength, high modulus articles involves the actual process of incorporating the fibers or fiber bundle into the matrix material to provide the desired end item. The methods heretofore proposed have included powder metallurgy techniques, liquid infiltration and plasma spraying. All of these techniques present problems, however.

In order to achieve the maximum strengthening effect in a fiber-reinforced article it is usually necessary to achieve a maximum volume of fiber. In the powder metallurgy and liquid infiltration processes, this maximum volume fill is necessarily not attainable since sufficient void volume must necessarily be provided around the individual fibers and between layers of fibers to provide for the requisite matrix infiltration. This is similarly true in the plasma spraying operations, although higher volume percentages are frequently more readily attainable by this method than by the infiltration techniques.

Furthermore, there is a serious reproducibility problem associated with these techniques which stems basicaly from an inability to insure that the matrix material will be provided in the same locations, in the same quantity and in the same quality in each article. Quite to the contrary, articles produced by the foregoing methods may vary significantly with respect to their physical properties despite the most careful attention to process details.

In most processes for forming the fiber-reinforced articles, of which winding may be taken as illustrative, particularly in high fiber volume applications, the individual filaments are laid up to closely abut or contact one another. As a consequence, it is usually impossible to provide complete peripheral encasement of each fiber using the pack, infiltration or spraying techniques. This is usual-

ly a fundamental requirement if the article is to have a strength approaching the calculated theoretical maximum, since the strength is a function of the filament-matrix bonding efficiency which is in turn a function of the contact area. Additionally, in the tightly-packed structures, it is virtually impossible to eliminate the void formation incident to incomplete coverage of the fibers with the matrix material. Of course the void formation can be minimized by a substantial overspacing of the fibers or by an overapplication of the matrix material, but both of these alternatives are inimical to the formation of a high filament volume article. The void formation is usually random both as to extent and location in structures formed by the foregoing methods and, hence, not only are the overall article strengths low, but the strengths and other characteristics are not uniform or predictable from one article to the next.

There are a number of other drawbacks to the use of the prior art processes which, while perhaps not as evident, are equally as detrimental to the overall character of the end product. The first drawback may be conveniently illustrated by reference to a particular composite filament consisting of boron with a thin, protective layer of stoichiometric silicon carbide. Such a filament is disclosed in a copending application entitled Composite Boron Filament by Malcolm Basche, Roy Fanti, and Salvatore F. Galasso, Ser. No. 618,513, filed Feb. 24, 1967, which shares a common assignee with the present invention.

Boron is one of the most promising filamentary materials. However, the reactivity of boron with a variety of metals, including those most suited to matrix applications, has not only limited the temperatures usable in the various fabrication procedures, but also limits the temperature at which the end products themselves are operational. The previously mentioned composite filaments avoid many of these problems, since the silicon carbide imparts a relative inertness to the boron as long as the coating is maintained intact, rendering the composite filament compatible with a wide variety of matrix materials.

In one typical use for these filaments, they are wound upon a mandrel in successive layers, the individual layers being subsequently infiltrated with the desired matrix material. Breakage of the filament in such a winding operation is a relatively common occurrence, particularly since short radius bends are possible where one fine filament is overlaid on another in a different winding direction. And even in operations where the composite filaments do not actually break, the short radius bend may rupture or stress the silicon carbide coating and, hence, render the boron substrate prone to degradation through a substrate-matrix interaction.

SUMMARY OF THE INVENTION

The present invention relates to fiber-reinforced articles and, more particularly, to an improved process for fabricating such articles to provide a maximum fiber fill, as desired, and impart reproducibility to the fabrication from one article to the next. It contemplates the use of composite filaments, particularly boron coated with silicon carbide, which are provided with an overcoat of a suitable matrix material, the coated composite filaments being formed into a desired shape and processed, as by hotpressing, into end products without the necessity for a supplementary matrix addition.

In a copending application entitled Composite Boron Filaments With Matrix Overcoat, by Malcolm Basche, Roy Fanti and Salvatore F. Galasso, Ser. No. 618,514, filed Feb. 24, 1967, which shares a common assignee with the instant application, there has been disclosed a composite filament comprising a boron substrate having a thin protective layer of stoichiometric silicon carbide and further provided with an overcoat of a suitable matrix material, including aluminum, magnesium, titanium and alloys and mixtures thereof. Not only are these fibers in some instances stronger than the basic fiber itself without the overcoat, but they are inherently less prone to damage and breakage in the production of the fiber-reinforced articles.

In accordance with one aspect of the present invention, the boron-silicon carbide composite filaments are provided with a matrix material overcoat of sufficient thickness to effect the desired filament-matrix ratio in the finished article; are wound or otherwise suitably disposed about or in a mandrel or mold of the correct configuration; and are pressed at a suitable temperature without the addition of additional matrix material to cause flow and bonding of the matrix material and the formation of a unitary article.

Because the matrix material is provided on and completely around each filament before and not during or after the winding operation, for example, the filament is less prone to breakage. Furthermore, the silicon carbide coating is less susceptible to rupture since some binding effect is provided by the matrix overcoat and since the effect of short radius bends is less pronounced. Still further, the ductility usually inherent in the character of the matrix tends to effect a load distribution over the fiber surface and minimizes stress concentrations therein, both mechanical and thermal.

The entire article fabrication process is greatly simplified and expedited since no intermediate operation is required to apply the matrix material. Since the correct filament-matrix volume ratio may be established prior to the article-fabricating process, variations in the ratio as a result of unavoidable variations in technique in the intermediate spraying operations, for example, are minimized. Furthermore, inasmuch as a complete filament encasement is provided, individual filament spacing is no longer critical in the sense that gapping must be provided for subsequent liquid infiltration. As a consequence, the maximum filament-matrix bond strength is achieved with the minimum variation in properties from one article to the next.

It is, of course, evident that in certain applications a maximum fiber fill is not necessary or desirable, for economic reasons perhaps. The article strength for a given situation may be achievable with less than the maximum fiber fill. While this may be accomplished by providing a thick matrix overcoat on the individual filaments, it may also be provided by utilizing supplementary infiltration or spraying of the matrix material, particularly between layers of filaments. Even in such cases, it will be found advisable to provide a substantial portion of the matrix material as an overcoat on the individual filaments. And whether or not a supplementary matrix material addition is utilized, adherence to the teachings herein will provide fiber-reinforced articles having improved properties including predictability of properties from one article to the next.

In another preferred aspect of the present invention, the basic filaments are provided with a matrix overcoat of substantial thickness but the compact is pressed under suitable pressure-temperature conditions such that complete elimination of the voids is not effected, although bonding between adjacent filaments is realized. Because careful control can be exercised over filament spacing and diameter, articles of closely controlled porosity may readily be produced, and in closely reproducible fashion. In this fabrication technique, a lesser pressure is utilized to form the structure, preferably without the addition of any additional matrix material, sufficient pressure being utilized to assure good contact between fibers for bonding, but less than that required to cause substantial flow of the matrix overcoat and complete filling of the voids.

It is, of course, understood that there is a pressure-temperature relationship involved in the flow and bonding mechanism described. In general, the higher the temperature involved in the fabrication operation, the less the pressure needed. And the particular parameters utilized will vary according to the materials utilized in the process. The specific conditions preferred in a given system will be evident to those skilled in the art.

The maximum fiber fill in a given article in the most closely packed structure, without fiber distortion, will be found to be 90.7 percent on an area basis. This dictates that the minimum matrix material overcoat, for the fiber-reinforced end product of maximum density, will be 9.3 percent. This maximum occurs in structures wherein the individual fibers are arranged in a close-packed triangular pattern. When other patterns are utilized a greater overcoat thickness will necessarily be required to insure complete elimination of the void spaced and, hence, to achieve the maximum fill.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Various experiments were conducted to establish the efficacy of the techniques heretobefore described. Silicon carbide-coated boron filaments were at various times provided with an overcoat of aluminum, magnesium and titanium, including such alloys at #2024 aluminum (nominal composition 4.5% copper, 1.5% magnesium, 0.6% manganese, balance aluminum) and various press and sinter techniques were utilized. In general, the fibers utilized have had a diameter of 3–5 mils with coatings provided thereover in various thicknesses, usually 0.1–0.2 mil.

The boron filaments were produced by chemical deposition on a heated ½ mil tungsten wire from a mixture of gases including boron trichloride and hydrogen, producing filaments in general having a diameter of 3–5 mils. A thin (0'2 mil) coating of stoichiometric silicon carbide was effected by chemical deposition on the heated boıon filament from a gas mixture including methyldichlorosilane, hydrogen and methane. Following the deposition of the silicon carbide, the filaments were provided with an overcoat of a ductile, low density material by plating, dipping and vapor deposition techniques. The particular overcoat materials utilized were sufficiently ductile to permit the transfer of tensile loads to the fiber by plastic deformation and included aluminum, magnesium, titanium and alloys thereof, as previously mentioned, the overcoat comprising, in general, 10 percent of the fiber or more on a cross-sectional basis when complete void elimination was described.

Example I

In one experiment, silicon carbide-coated boron filaments provided with an overcoat of pure aluminum were packed in a short Pyrex tube having an 0.118 inch outer diameter and a 0.078 inch internal diameter. The tube was heated at its central portion to approximately 600° C. which is just below the melting point of aluminum and stretched to provide a necked-down portion in the tube having an outer diameter of 0.0375 inch and a resultant internal diameter of 0.0250 inch. A subsequent examination of sections taken through the necked-down portion revealed a complete absence of voids.

Example II

In another experiment, silicon carbide-coated boron filaments with an aluminum overcoat were hot pressed in a mold at 10,000 p.s.i. at a temperature of 500° C. The mold was heated initially to 550° C. and the temperature was allowed to decay to 500° C. before the pressure was applied. Analysis of the flat plate produced revealed the total absence of voids in these compacts.

It will readily be seen that, through the use of the techniques hereinbefore described, fiber-reinforced articles of carefully controlled density or maximum theoretical density can readily and reproducibly be fabricated.

While the invention has been described with reference to specific examples, fabrication parameters and materials, these embodiments and conditions are intended to be illustrative only. Various modifications and alternatives will be readily evident to those skilled in the art within the true spirit and scope of the invention as set forth in the appended claims.

What is claimed is:

1. In the processes for forming fiber-reinforced articles utilizing composite filaments of silicon carbide-coated boron which have been provided with an overcoat of a ductile matrix material, the improvement which comprises:
   providing the overcoat on the individual filaments in the filament-matrix volume ratio desired in the finished article;
   disposing the filaments in a predetermined relationship with one another;
   forming the filaments into a compact of the desired shape without the addition of supplementary quantities of matrix material under a pressure sufficient to effect the formation of a compact having a predetermined density;
   and applying sufficient heating in the process to effect a substantially complete metallurgical bonding of the matrix material between adjacent filaments and the formation of a unitary article.

2. In the processes for forming fiber-reinforced articles utilizing composite filaments of silicon carbide-coated boron which have been provided with an overcoat of a ductile matrix material, the improvement which comprises:
   providing the overcoat on the individual filaments in the filament-matrix volume ratio desired in the finished article;
   disposing the filaments in a predetermined relationship with one another;

   forming the filaments into a compact of the desired shape without the addition of supplementary quantities of matrix material under a pressure sufficient to cause flow of the matrix material and the formation of a compact having a density approximating the maximum theoretical density;
   and applying sufficient heating in the process to effect a substantially complete metallurgical bonding of the matrix material between adjacent filaments and the formation of a unitary article.

3. The improvement according to claim 2 in which: the overcoat is provided to a thickness comprising at least 10 percent of the filament area on a cross-sectional basis.

4. The improvement according to claim 3 in which: the matrix material is selected from the group consisting of aluminum, magnesium, titanium and alloys therof.

5. The improvement according to claim 4 in which: the pressure and heating are simultaneously effected in the formation of the compact.

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