United States Patent

[11] 3,571,901

[72] Inventor Raymond Vincent Sara
North Olmsted, Ohio
[21] Appl. No. 833,009
[22] Filed June 13, 1969
Division of Ser. No. 727,898, May 9, 1968.
[45] Patented Mar. 23, 1971
[73] Assignee Union Carbide Corporation
New York, N.Y.

[54] **METHOD OF FABRICATING A CARBON-FIBER REINFORCED COMPOSITE ARTICLE**
5 Claims, 2 Drawing Figs.

[52] U.S. Cl. 29/419, 29/527.3, 29/527.5, 164/80, 164/91
[51] Int. Cl. B23p 17/00
[50] Field of Search 164/80, 91, 97; 29/182.2, 182.5; 29/419, 527.3, 527.5

[56] **References Cited**

UNITED STATES PATENTS

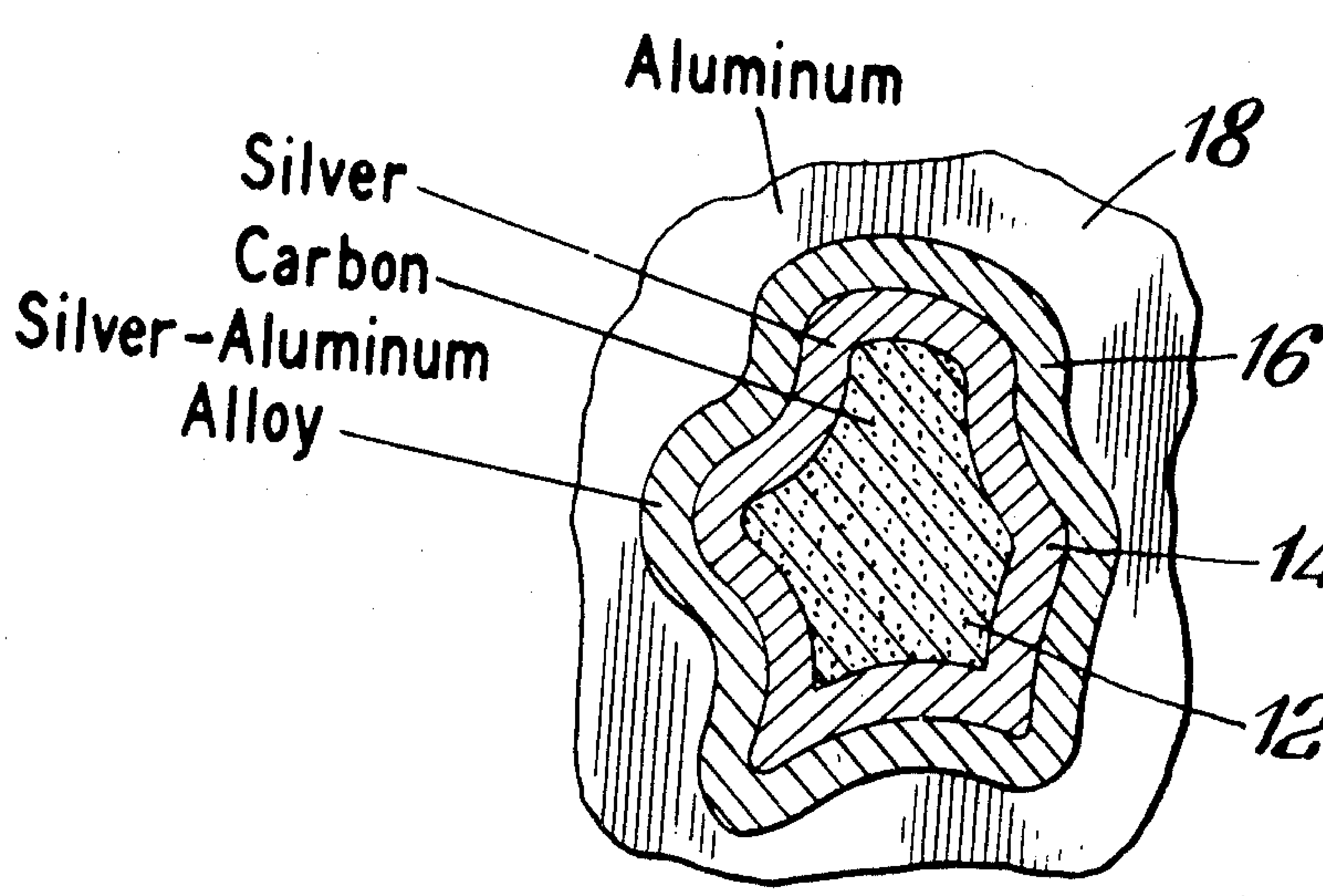
910,674	1/1909	Hancock	164/86
2,937,436	5/1960	Butler et al.	29/419
3,038,248	6/1962	Kremer	29/419
3,187,422	6/1965	Morgan	29/419
3,256,596	6/1966	Fiedler	29/419
3,476,529	11/1969	Dubin et al.	117/71X
3,491,055	1/1970	Talley	260/37

OTHER REFERENCES

" Metal-Ceramic Composite Structural Materials" L. N. Hjelm S.A.M.P.E. Journal, Nov., 1965.

Primary Examiner—John F. Campbell
Assistant Examiner—D. C. Reiley
Attorneys—Paul A. Rose, Robert C. Cummings, Frederick J. Mc Carthy, Jr. and John S. Piscitell

ABSTRACT: Light weight composite refractory articles comprising a plurality of metal or alloy coated carbon fibers bonded together by an aluminum base matrix are provided. The coating metal or alloy can be any material selected from the group consisting of silver, silver-aluminum base alloys and mixtures thereof. The preferred process for producing such composites comprises contacting silver coated carbon fibers with an aluminum base material followed by hot pressing the so-contacted fibers at the solidus temperature of the aluminum base material to infiltrate aluminum around the silver-coated fibers and then cooling the resultant article to a completely solidified state. Such composite articles are characterized by a high tensile strength coupled with a high modulus of elasticity and are especially suited as materials of construction for aerospace and hydrospace vehicles and systems.



PATENTED MAR 2 3 1971 3,571,901

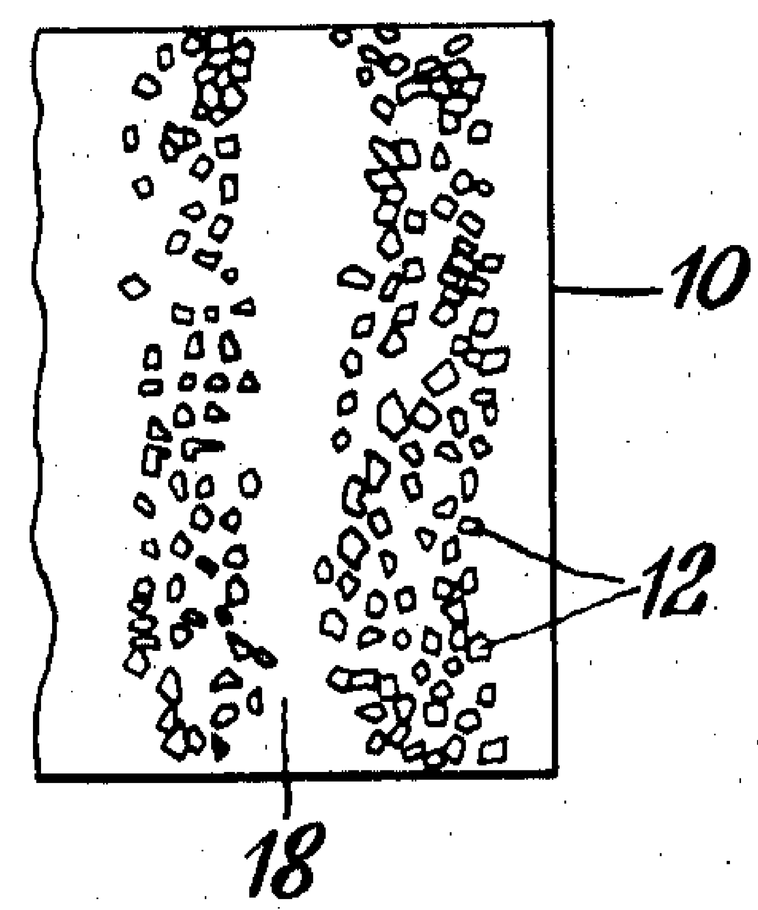


Fig. 1.

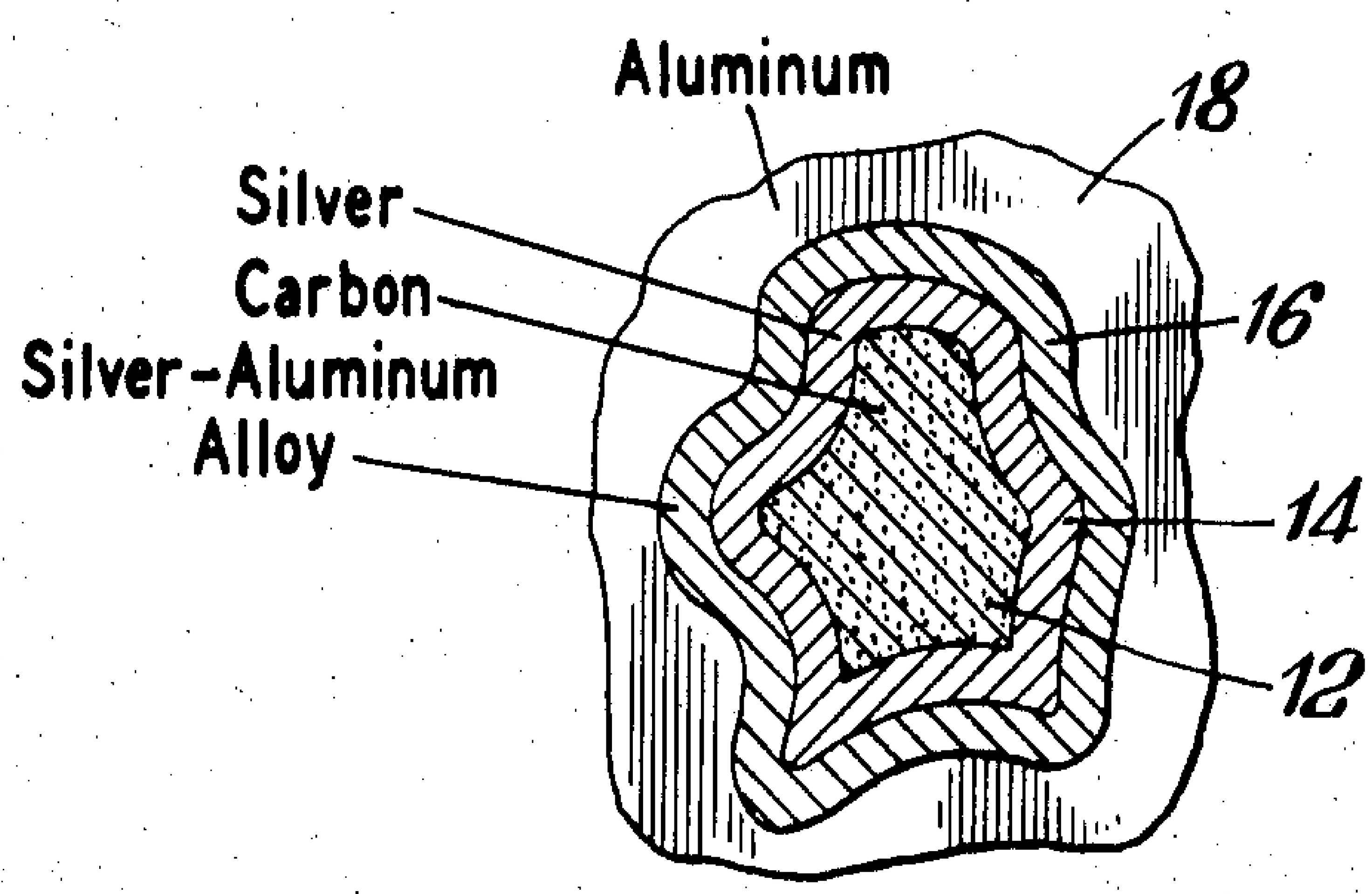


Fig. 2.

INVENTOR.
RAYMOND V. SARA

BY John S. Piscitello

ATTORNEY

METHOD OF FABRICATING A CARBON-FIBER REINFORCED COMPOSITE ARTICLE

This application is a division of copending application Ser. No. 727,898, filed May 9, 1968.

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to high strength, high modulus composite refractory articles composed of a plurality of carbon fibers which are essentially coated with a thin layer of silver and/or silver base alloys and bonded together by an aluminum base matrix and to the process of producing the same.

2. Description of the Prior Art

In the space and missile industries, there has developed a need for refractory materials of construction which exhibit exceptional physical properties, for example, low density coupled with high strength and stiffness. Attempts to produce such refractory materials have centered around the fabrication of composite articles.

One of the most promising materials available today for use in composite form is carbon textiles since they possess excellent refractory properties and are commercially available in all known textile forms. Today, it is well known to form composites of carbon textiles and resins.

Recently, efforts have been directed to forming composites of carbon textiles and metals. The prime object of forming such composites is to increase the strength of the metal matrix by the inclusion therein of high strength carbon fibers.

Aluminum has been suggested as the matrix media for carbon fiber-metal composites which are intended for use in aerospace applications, in the main, due to its low density. However, attempts heretofore to incorporate carbon fibers in an aluminum matrix have met with little or no practical success due to the fact that carbon, especially its graphic form, is not readily wetted by molten aluminum.

The present invention overcomes the foregoing problem by employing an intermediate coating or wetting agent which when applied to the carbon fibers enables them to be readily bonded together by an aluminum base matrix material.

SUMMARY

Broadly stated, the carbon fiber-aluminum composite article of the invention comprises a plurality of carbon fibers each of which is coated with either a thin layer of silver, a silver-aluminum base alloy or mixtures thereof and bonded together, preferably in a side-by-side or parallel manner, with an aluminum binder or matrix material. Generally, this composite article may be provided by a process which comprises coating carbon fibers with a thin but essentially continuous film of silver, contacting the so-coated fibers with a solidified aluminum base material, hot pressing the so-produced assembly at the solidus temperature of the aluminum base material to infiltrate it around the coated fibers and cooling the resultant aluminum bonded carbon fibers to produce a composite article. This article can then be formed into any desired shape by known techniques which will readily suggest themselves to those skilled in the art.

DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagrammatical illustration of a portion of a carbon fiber-aluminum composite article produced according to the preferred teachings of the invention; and

FIG. 2 is a greatly magnified diagrammatical illustration of a single fiber found in the composite of FIG. 1.

Referring now to the drawings in detail, FIG. 1 shows a portion of a rectangular composite article **10** consisting of aligned graphite fibers **12** (the silver coating and aluminum-silver zone thereon are not shown) bonded together by an aluminum matrix **18**. The graphite fibers **12** are disposed in the aluminum matrix in a parallel manner with their length dimension being perpendicular to the surface of the drawing. FIG. 2 shows a single graphite fiber **12** taken from the composite **10** of FIG. **1** having on its surface a coating of silver **14** and an aluminum-silver interface zone **16** between the silver coating and the aluminum matrix **18**.

DESCRIPTION OF THE PREFERRED EMBODIMENT OF THE INVENTION

Carbon textiles in any form can be employed in the practice of the instant invention. However, it is preferred to employ carbon fibers in yarn or multifilament form. Carbon textiles are available commercially and are generally produced by the techniques described in U.S. Pat. Nos. 3,107,152 and 3,116,975, among others.

Silver can be deposited on the carbon fibers by a variety of methods. The techniques available for accomplishing this include electrodeposition from a conductive bath, reduction of salts to the metal or sputtering. The exact deposition technique to be employed is dictated by a number of factors. Sputtering can be used on relatively complex shapes and results in a tenacious bond between the thin silver coating and the carbon fiber substrate. Such a bond is a highly desirable feature in carbon fiber-metal matrix composites. Dipping of the carbon fibers into a silver containing solution and chemically reducing the salt to the metal can also be employed, however, with this technique it is difficult to accurately control the thickness of the silver coating. Electrodeposition of silver from a conductive bath is an ideal way of coating carbon fibers with a thin film of silver and is the preferred method since it results in a uniform, tenaciously bonded metallic coating.

The following example illustrates in detail the preferred practice of the instant invention.

A single ply graphite yarn composed of 720 monofilaments, each characterized by an average modulus of elasticity of 33.5 $\times 10^6$ lb./in.^{2} and an average tensile strength of 20 $\times$ **10**^{4} lb./in.^{2}, was cut into a plurality of 4inch lengths. These lengths of yarn were heated at 560° C. for 30 seconds in a tube furnace which was open to the atmosphere. The slightly oxidized fibers were immediately plated electrolytically with silver by placing them in a strike solution for about 5 to 10 seconds and then into a conventional plating solution for about 5 to 10 seconds. The plating current employed was about 400 milliamps for the strike solution and between about 600 to 800 milliamps for the conventional plating solution. The make up of the strike and conventional plating solution was as follows:

Strike Solution

Silver Cyanide (Ag CN) 6.5 g./l.
Potassium Cyanide (KCN) 68.0 g./l.

Regular Solution

Silver Cyanide (Ag CN) 41 g./l.
Potassium Cyanide (KCN) 40 g./l.
Potassium Hydroxide (KOH) 11 g./l.
Potassium Carbonate (K_2CO_3) 62 g./l.

The so-coated graphite fibers were cut into one-inch lengths and then positioned in a parallel manner between alternate layers of aluminum alloy foils measuring 0.003 inch $\times$ 0.125 inch $\times$ 1.00 inch each. The aluminum alloy employed consisted of 4.5 weight percent copper, 0.6 weight manganese, 1.5 weight percent magnesium with the remainder being essentially aluminum. Two single plies of silver-coated yarn were used for each layer. The complete array consisted of 22 layers of foil and 42, one-inch lengths of silver-coated single ply yarn. This assembly was placed in a graphite mold and hot pressed at 2,250 lb./in.^{2} in vacuum at approximately 550° C. for one hour. The composite was subsequently cooled in the mold to room temperature and then formed into a 1/16″ ×⅛″ × 1″ specimen for physical property measurements. This specimen evidenced an average tensile strength of 44,100 lb./in.^{2} and a Young's modulus of 13.0 $\times 10^6$ lb./in.^{2}.

The table below shows the improvement obtained by incorporating silver coated graphite fibers in an aluminum base matrix material by the technique of the instant invention.

TABLE

PROPERTIES OF GRAPHITE-FIBER, ALUMINUM COMPOSITE

Aluminum Layers in Composite		Volume Percent of Fibers	Young's Modulus $\times 10^6$ lb/in^2	Tensile Strength lb/in^2
o	Matrix Metal	0	10.4	36,100
24	Silver-Coated Fibers	16	13.0	44,100
30	Silver-Coated Fibers	28	15.3	56,700

Composites produced by the technique of the instant invention are extremely useful as materials of construction for subsonic and supersonic aircraft, space system components and various propulsion devices.

It will be appreciated by those skilled in the art that while the preferred technique of the invention results in a composite article which comprises silver-coated carbon fibers bonded together by an aluminum base matrix, the silver coating on the carbon fibers may be converted to a silver-aluminum base alloy by annealing the composite during its fabrication or after it has been formed. The only requirement being that the silver-aluminum base alloy not be formed at a temperature in excess of the melting point of the applied silver coating. Accordingly, it is clear that the instant invention results in a unique type of composite comprising carbon fiber which are coated with either silver, a silver-aluminum base alloy or mixture thereof and bonded together by an aluminum base matrix.

While the foregoing example concerns a composite having the fibers positioned in a side-by-side relationship, it is readily apparent to those skilled in the art that the carbon fibers may be randomly orientated in the aluminum matrix if more isotropic physical properties are desired without losing the benefits of the instant invention. In addition, it is obvious that the thickness of the silver can be varied as desired. All that is required is that it be thick enough to prevent the aluminum matrix metal from coming into contact with the reinforcing carbon fibers to such an extent that there is essentially no bonding between the fibers and the matrix metal. Likewise, it will be appreciated by those versed in the art that although graphite fibers and fabrics are preferred in the practice of the instant invention, nongraphitic carbon fibers and fabrics may also be employed.

It should be noted here that while the invention can be beneficially practiced by using pure aluminum as the matrix metal, it is preferred to employ an aluminum base alloy which has an incongruent melting temperature. Such alloys are well known in the art and will not be discussed herein in detail.

In addition it should be noted that while it may be possible to infiltrate the molten aluminum metal around the silver clad carbon fibers without removing the silver therefrom, the preferred practice of the invention involves infiltrating the matrix metal around the filaments by heating the metal to a temperature at which it begins to melt but below that at which it becomes completely liquid, i.e., at its solidus temperature,

and then forcing the so-softened matrix metal around the individual filaments by the means of an applied pressure. The amount of pressure to be applied is not particularly critical and need only be sufficient to compact the fibers and matrix material into an essentially nonporous article.

The term carbon as used herein and in the appended claims is meant to include both the nongraphitic and graphitic forms of carbon.

The foregoing example is presented for illustrative purposes only and is not intended to unduly limit the reasonable scope of the instant invention. The limitations of applicant's invention are defined by the following claims.

I claim:

1. A method of producing a metal coated carbon fiber aluminum-bonded composite article comprising:
   a. contacting a plurality of silver coated carbon fibers with a solidified aluminum base material;
   b. heating the so-formed mass to the solidus temperature of the aluminum base material and exerting a force thereon sufficient to cause said aluminum base material to be infiltrated around said silver coated fibers; and
   c. cooling the resultant composite to a temperature at which said aluminum base material completely solidifies to form an aluminum-bonded silver-coated fiber composite article.

2. The method of claim 1 wherein said aluminum base material is an alloy consisting essentially of 4.5 weight percent copper, 0.6 weight percent manganese, 1.5 weight percent magnesium with the remainder being essentially aluminum.

3. A method of producing an aluminum-bonded silver-coated carbon fiber composite article comprising:
   a. forming an assembly comprising alternate layers of silver coated carbon fibers and aluminum base foil, said fibers being positioned thereon in a parallel manner;
   b. placing the so-formed assembly in mold and heating it in an inert atmosphere to a temperature approximately equal to the solidus temperature of said aluminum base foil while exerting a sufficient pressure on said assembly to cause said aluminum base foil material to be infiltrated around said silver-coated carbon fibers; and
   c. cooling the resultant assembly to produce an aluminum-bonded silver-coated carbon fiber composite.

4. The method of claim 3 wherein:
   a. said aluminum base foil material consists essentially of 4.5 weight percent copper, 0.6 weight percent manganese, 1.5 weight percent magnesium with the remainder being essentially aluminum; and
   b. said heating in step (b) is accomplished in a vacuum at a temperature of about 550° C.

5. The method of claim 1 wherein said aluminum-bonded silver-coated carbon fiber composite article is heated to a temperature sufficient to cause at least some said silver to alloy with at least some of said aluminum to form at least a partial coating of a silver-aluminum base alloy on said fibers.