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PROCESS OF PRODUCING CARBONIZED ARTICLES

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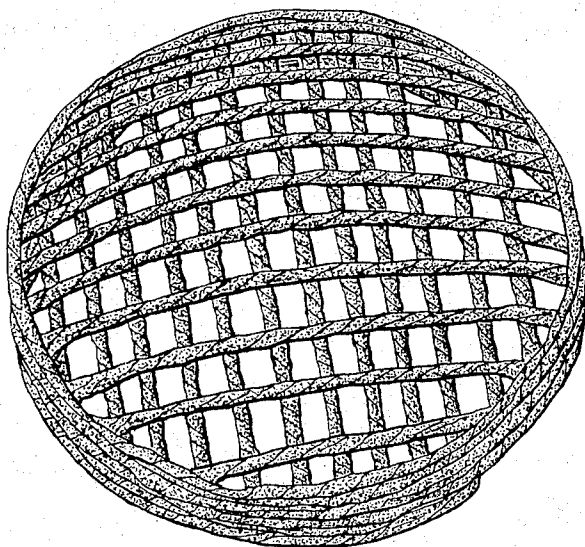


Fig. 1.

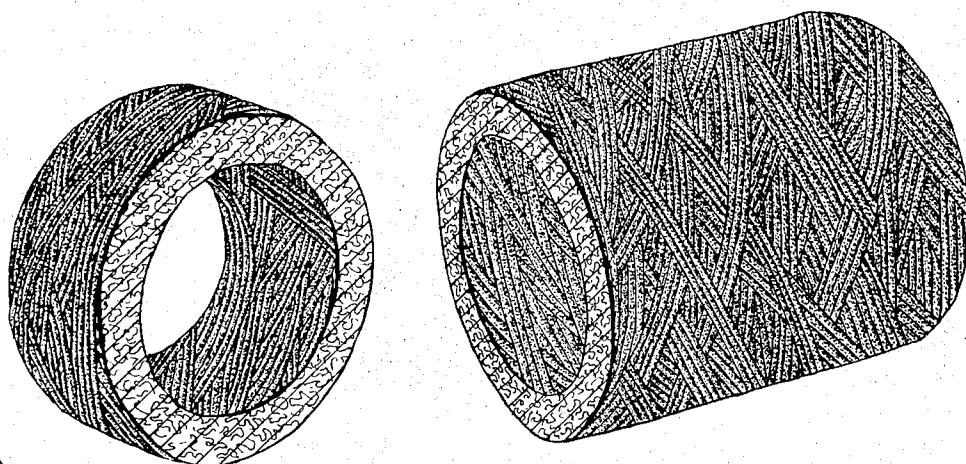


Fig. 2.

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PROCESS OF PRODUCING CARBONIZED ARTICLES

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This invention relates to a method for fabricating carbonaceous articles, and more particularly to the fabrication of intricately shaped carbonaceous articles.

Rigid, thin walled carbonaceous articles possessing intricate predesigned shapes can presently only be formed or machined from conventional types of carbon or graphite with an excessive amount of breakage.

The object of this invention is to provide a process for manufacturing carbonaceous articles of an intricate shape, which process avoids the disadvantages of the prior art.

It is also the object of this invention to provide a process whereby a designer has more freedom to tailor a carbonaceous body to meet size, shape and specific design requirements.

Broadly stated, the objects of the invention are accomplished by a process which comprises the formation of carbonaceous articles from flexible carbonaceous fibers, the carbonaceous fibers being treated with a coke depositing resin binder such as by being coated or impregnated with the binder, and subsequently curing said binder to form a rigid carbonaceous article which is infusible and has structural integrity at high temperatures.

As used herein and in the appended claims, the term "flexible carbonaceous fibers" refers to materials which have the physical properties of a textile, such as hand and drape, coupled with at least in part, the attendant chemical and electrical properties of carbon. Such materials include, but are not intended to be limited to the following:

(a) The heat treated material which is disclosed and claimed in co-pending U.S. patent application, Ser. No. 224,989, filed Sept. 20, 1962, now Patent No. 3,305,315, one of the co-inventors named in such application being the inventor in the subject application. Briefly stated, heat treated material is defined as a non-flammable, non-fusible, strong and flexible material of cellulosic origin, which has undergone an approximate weight loss based on the starting cellulosic material in the range of from about 20% to about 50% which has a volatile content of approximately 45% to 70% and yields a fixed carbon residue of approximately 30% to 55% by weight when determined by a standard procedure (ASTM-D 189-61) (Standard Method of Test for Conradson Carbon Residues of Petroleum Products). This material may be manufactured by a controlled thermal treatment comprising heating of the cellulosic starting materials at a temperature of up to about 350° C. Such thermal treatment usually is performed in the presence of an oxidizing atmosphere and acid. When acid is employed, the thermal treatment is done in the range from about 100° C. to 350° C., and when acid is not used, the thermal treating is performed in the temperature range from about 180° C. to 350° C.

(b) The manufactured textile carbon which is disclosed in U.S. Patent 3,011,981, issued Dec. 5, 1961, to W. T. Soltes. It is disclosed in said patent that a textile carbon which retains the flexibility and other physical characteristics of the textile starting material may be provided by the thermal conversion of fibrous and substantially pure cellulosic materials such as strands, skeins, ropes, cloths, fabrics, and batting pads; and

(c) The electrically conductive graphite in a flexible fiber and fabric form which is reported in Metal Progress, May 1959, pp. 115-116, and which is commercially avail-

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able in any textile form such as yarns, braids, felts and woven or knit fabrics, such as cloth.

The particular flexible carbonaceous material to which this invention is directed are carbonaceous fibers having a diameter of about 5 to 100 microns and which are arranged in a parallel pattern such as yarn or tow.

Any of the conventional carbonizable binders which are well known in the art, both of the thermoplastic (e.g. soft and medium pitch) variety and the thermosetting (e.g., phenolic resins, epoxides, and furfural-furfuryl alcohol mixtures) variety are suitable in the practice of the invention.

The binder employed in the practice of the invention may be applied to the carbonaceous fibers before or after the desired article is shaped. Once the article is shaped and the binder applied, the binder is cured in the manner which is conventional for the particular selected binder, i.e., by chemical catalyst or by thermal treatment, with or without pressure as needed. The best method of curing a particular binder will be readily apparent to those skilled in the art. Subsequent to the curing operation, the cured binder may be carbonized or graphitized if desired in a conventional manner.

Among the suitable techniques for shaping the articles of the invention are shaping procedures which utilize a fusible form or mold and filament winding techniques.

The invention will be made readily understood by reference to the accompanying drawing wherein:

FIG. 1 is an isometric view of an intricately shaped carbonaceous article of the invention which has been shaped by utilizing a fusible form, and

FIG. 2 is an isometric view which is broken away to show a cross-section of another uniquely shaped carbonaceous article of the invention which has been shaped by filament winding.

Referring to FIG. 1, a rigid, intricately shaped article having both open spacing in the form of a grid and a wound rim is shown.

This article was formed as follows:

Example I

A lead form was made having the desired diameter and its dome was provided with a grid-like pattern. Pieces of graphite yarn which were wet with a binder comprising by weight 25 percent furfuryl alcohol, 25 percent furfural and 50 percent phenol formaldehyde resin were laid out in the grid of the dome in the illustrated configuration and the rim of the article was formed by winding a continuous piece of graphite yarn also wet with the same binder in parallel layers around the ends of the grid pieces. A rigid article was then formed by curing the binder at 150° C. Subsequently the fusible lead form was melted out and the article was baked to 900° C.

Thus, the finished article consisted of oriented graphite fibers bonded together with a carbonized binder. This article has been found to withstand rough handling and will not break or chip when dropped. A similar article machined from conventional graphite would have egg-shell properties.

Referring now to FIG. 2, a cylinder which is formed of carbonaceous fibers which are disposed in overlapping helical pitched layers is shown. The article of FIG. 2 was provided by winding carbonaceous fibers which had been wetted with a binder comprising by weight 25 percent furfuryl alcohol, 25 percent furfural and 50 percent phenol formaldehyde resin on a form which was subsequently removed. The binder was then cured by heating to approximately 150° C. to provide a rigid cylinder.

In articles of the type of FIG. 2, the fibers may be pitched at any angle to one another that the designer selects in order to obtain optimum stress relationships.

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It will be appreciated from the above, that by the process of the invention, namely combining oriented carbonaceous fibers with a carbonizable coking binder, a whole new family of carbonaceous articles is possible. Such articles, not only are more fracture resistant, more resilient and more chip resistant than conventional carbon or graphite shapes, but they may be formed to meet specific design requirements such as size, shape, stress relationships and other design criteria. Thus, any shape in which yarn or tow can be wound, stacked or laid out in a pre-selected pattern may, by the practice of the invention, be provided in carbonaceous form. In addition an integral combination of carbonaceous forms can be made, i.e., layers of heat treated material and/or carbon and/or graphite can be built into one article. This would permit optimum usage of shrinkage, thermal conductivity, strength, price and other properties commonly distinguished between forms of elemental carbon.

I claim:

1. A process for manufacturing a formed carbonaceous article which comprises disposing parallel oriented carbonaceous fibers of cellulosic origin having a volatile content of between 45 and 70 percent by weight in a pre-selected pattern and subsequently subjecting said article after said fibers have been treated with a carbonizable binder to a carbonizing operation to thereby form a rigid article.

2. The process of claim 1 wherein said carbonizable binders are selected from the group consisting of thermoplastic binders and thermosetting binders.

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3. The process of claim 2 wherein said fibers are disposed on a fusible mold in a preselected pattern.

4. The process of claim 1 wherein said article is subsequently subjected to a graphitizing temperature.

5 5. The process of claim 3 wherein said fibers have been treated with a binder comprising 25 percent furfuryl alcohol, 25 percent furfural and 50 percent phenol formaldehyde resin, and the fusible form subsequently removed by heating at an elevated temperature.

10 6. The process of claim 5 wherein said article is subsequently subjected to a graphitizing temperature.

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