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LOW TEMPERATURE METHOD FOR PRODUCING AMORPHOUS BORON-CARBON DEPOSITS

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ABSTRACT OF THE DISCLOSURE

Process for producing an amorphous boron-carbon deposit on a filamentary substrate at a temperature below 1,000° C. by heating the filamentary substrate in a vacuum chamber to a temperature above 700° C., preferably in the range 750–850° C., and contacting it with a hydrocarbon-substituted borane gas such as ethyl decaborane or triethyl borane.

This invention relates to a process for producing amorphous boron-carbon deposits and more particularly to a process in which amorphous boron-carbon deposits are produced at relatively low temperatures.

Amorphous boron-carbon deposits, either in the boron carbide form or in non-stoichiometric mixtures of boron and carbon, have been found to have highly desirable mechanical propertiers. In particular, these materials are known for their extremely high tensile strength and modulus of elasticity even at very high temperatures. It should be noted that while this material is referred to herein as "amorphous," this term is not intended to signify the complete absence of a crystalline structure in the deposit but instead indicates only that no crystalline structure is discernible by the presently available techniques, such as X-ray diffraction, etc.

Despite the highly desirable mechanical properties of amorphous boron-carbon deposits, utilization of these materials has been limited generally by the fact that prior art processes for producing such materials have required process temperatures which would destroy or cause degradation of many substrate materials. Further, these processes have been relatively slow, and have not been capable of consistently producing high quality deposits.

For example, the most common method for producing amorphous boron-carbon deposits heretofore has been to heat the substrate to temperatures above 1,100° C. in the presence of boron trichloride, methane, and hydrogen. Substrates which undergo undesirable physical or chemical changes in this temperature range cannot be used in this process.

It is an object of the present invention therefore to provide a method for producing amorphous boron-carbon deposits at a relatively low temperature.

Another object of this invention is to provide a practical low temperature process for producing amorphous boron-carbon deposits in a form which can be utilized as a structural material reinforcement.

These and other objects are met, in accordance with the present invention, by a process which comprises, briefly, heating a substrate material to a temperature in the range from 700–900° C. at a very low pressure and contacting the substrate material with a hydrocarbon-substituted borane gas. In the preferred form of the present invention a filamentary substrate is contacted with an alkyl-substituted borane at a temperature of 750–850° C. and a pressure of from 10–25 torr.

While the specification concludes with claims particularly pointing out and distinctly claiming the subject matter of the present invention, this invention may be better understood from the following description.

With regard generally to the substituted boranes which constitute the source of both boron and carbon in the present invention, a variety of hydrocarbon-substituted boranes maybe useful but the preferred materials are the lower alkyl-substituted boranes, such as ethyl decaborane and triethyl borane. These materials are relatively stable at ordinary temperatures and apparently undergo controlled decomposition at the reaction temperature and pressure of the present process. Although the specific mode of decomposition is unknown, it is likely that numerous intermediate products are formed as the decomposition proceeds.

The present invention generally requires a very low pressure in order to effect controlled decomposition and to minimize extraneous side reactions. More specifically, the pressure at which the process of the present invention is conducted should be below 25 torr. The pressure range from 10 to 25 torr is preferred. It should be noted that a torr is equal to a pressure of 1 millimeter of mercury, absolute.

The objects of the present invention make it clear that this invention is directed to a low temperature process. Generally, this is a process in which amorphous boron-carbon deposits are produced on substrates which need not be heated in excess of 1,000° C. Although substrate temperatures throughout the range from 700–900° C. have been found to be effective to produce amorphorus boron-carbon deposits in accordance with the present invention, temperatures in the range from 750–850° C. are preferred.

The effectiveness of the present invention to produce deposits in the temperature range below 1,000° C. is an important feature of the process taught herein. It permits the production of amorphous boron-carbon deposits on many substrates which would undergo undesirable physical or chemical changes at temperatures below 1,000° C. The latter category includes metallic substrates, which may undergo crystalline changes or exhibit a loss in ductility if they are heated above 1,000° C., and also includes substrates such as silica which would deteriorate at temperatures above 1,000° C. The latter is a particularly desirable substrate for use in producing low density boron-carbon filaments. Such filaments, produced, in accordance with the present invention, by the deposition of amorphous boron-carbon on a filamentary silica substrate, have excellent potential as the reinforcing constituent of composite structural materials.

The quality of these products has been determined by visual observation as well as by microscopic analysis which showed the deposits to be relatively smooth and free of imperfections.

As an example of the present invention, an experiment was conducted in which a tungsten filament, 1 mil in diameter, was mounted in a tubular quartz reactor having an internal diameter of ¾ inches and a length of 2 inches. Inlet and outlet tubes were connected to the reactor, mounted at right angles to the tubular axis of the reactor. The outlet tube was connected to a vacuum-producing apparatus and the inlet to a source of triethyl borane. When the pressure had been reduced to about 18–22 torr, and electrical contacts in the tubular reactor had heated the tungsten filament to about 900° C., triethyl borane was admitted to the reactor at a relatively high rate. For thirty minutes, the triethyl borane was continuously passed through the reactor while the pressure in the reactor was held at 18–22 torr. During this time, the filament had grown to a diameter of 3.27 mils, roughly at a diameter growth rate of 4.5 mils per hour. Later, this experiment was repeated at a lower temperature, about 700–800 C., and at a slightly higher feed rate of triethyl borane. The pressure in the latter experiment was held at 13–15 torr. In this later experiment, a filament 2.64 mils in diameter

was produced in 13 minutes at a diameter growth rate of 8 mils per hour.

In still another example of the present invention, a low temperature filamentary substrate, specifically silica, was used. The silica substrate was 0.64 mil in diameter and had a thin carbon coating on its surface. Again, the substrate filament was resistively heated to 700–800° C., and the pressure was held at 13–15 torr. In this example, a deposition rate of about .7 mil per hour was attained. In a modification of this example, another experiment was conducted in which ethyl decaborane was contacted with a silica substrate at 800–850° C. This also resulted in the deposition of amorphous boron-carbon on the silica filament.

The filaments produced in all the above examples were typically hard and, under X-ray analysis, devoid of any discernible crystal structure. In order to ascertain that the deposits were not pure boron, an attempt was made to etch these deposits with 50% hydrogen peroxide at 100° C. No etching occurred, which indicated that the deposits were not simply boron. Since carbon would not be expected to be deposited at the low temperatures of these experiments, it is also unlikely that the deposits produced were simply carbon. Therefore, it was concluded that the deposits produced in these experiments were amorphous boron-carbon deposits. A portion of these deposits may have been boron carbide but this could not be proven.

Since high quality, low density amorphorus boron-carbon filaments are likely to be highly desirable as the reinforcement constituent of composite structural materials, an adaptation of the present process to some form of continuous deposition apparatus will probably be required to provide such filaments in the quantities required. In one such adaptation demonstrating the practicality of a continuous form of the present invention, the filamentary substrate was passed continuously through the length of a tubular reactor in which it was heated and exposed to a continuously flowing hydrocarbon-substituted borane. An amorphous boron-carbon filament was obtained.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. A process for producing a high strength filament consisting essentially of an amorphous boron-carbon deposit, said process comprising placing a filamentary substrate material in a vacuum chamber, evacuating said chamber to a pressure below 25 torr, heating said substrate material to a temperature in the range 700–900° C., and contacting said substrate material with ethyl decaborane.

2. A process, such as that recited in claim 1, wherein said chamber is evacuated to a pressure in the range from 10 to 25 torr.

3. A process, such as that recited in claim 1, wherein said process operates continuously on a substrate material of indefinite length which passes through said vacuum chamber after said chamber has been evacuated and as said gas is also passed continuously through said vacuum chamber.

4. A process, such as that recited in claim 1, wherein said temperature is in the range from 750–850° C.

5. A process, such as that recited in claim 1, wherein said substrate material is filamentary silica.

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