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MICROWAVE SINTERING OF BORON CARBIDE

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BACKGROUND OF THE INVENTION

This invention relates to materials technology. More particularly, it relates to the preparation of ceramic materials. This invention is the result of a contract with the Department of Energy (Contract No. W-7405-ENG-36).

Boron carbide is an unusually hard and wear resistant ceramic material which is refractory and has a low specific weight. It has numerous diverse uses, including tooling for metal-working machinery, sand blasting nozzles, armor for military vehicles, as neutron absorption materials, and structural applications.

Conventional densification of boron carbide is accomplished by hot-pressing or pressureless sintering. To obtain high densities, it has been necessary to mix a densifying aid, usually carbon, with the starting material, boron carbide powder. Other densification aids which have been used include titanium diboride and aluminum trifluoride. The amount of densification aid required varies from about 2 to about 8 wt%. In order to achieve densities of 95% of theoretical density and above, a densification aid and a temperature in excess of about 2150°C is required. One group of researchers was able to achieve a density of only 87% of theoretical upon heating to 2100°C without carbon additions. With carbon addition of 3%, they were able to achieve densities as high as 98% of theoretical by heating to 2150°C.

SUMMARY OF THE INVENTION

This invention is a method for forming boron carbide into a particular shape and densifying the green boron carbide shape. Boron carbide in powder form is pressed into a green shape and then sintered, using a microwave oven, to obtain a dense boron carbide body, or workpiece.

More specifically, in a broad embodiment, the inventive method comprises: pressing boron carbide powder to form a workpiece; exposing said workpiece to microwave radiation in an inert atmosphere until a peak temperature is reached; holding said workpiece at about said peak temperature for a time period; and cooling said workpiece in an inert atmosphere.

There are several advantages to using microwave heating, which may be described as volumetric, that is, heating is uniform throughout the volume of the workpiece, because microwave energy is coupled to the whole of the workpiece. This is in contrast to heating in a conventional oven, where the interior of a workpiece is heated by conduction from the surface of the workpiece. Better mechanical properties result from the more uniform microstructure achieved because heating is volumetric. Thermal stresses are minimized because of the homogenous nature of volumetric heating; this will allow the fabrication of more complex shapes than are now possible by conventional processing. Also, the use of microwave heating results in shorter processing times and lower energy costs.

DETAILED DESCRIPTION OF THE INVENTION

In the experimentation described herein, the starting material was boron carbide powder sold by ESK of Munich, West Germany under the trade name Tetrabor 1500®. The average particle size of the powder is 1.1 μm and the B.E.T. surface area is 12 m^2/gm.

The microwave oven used in the experimentation has a 2 ft X 2 ft X 2 ft (0.6 m X 0.6 m X 0.6 m) resonant cavity equipped with a rotating specimen table and a two-color infrared pyrometer. The oven operates at 2.45 GHz and has a 6 kW power supply. Various power levels were used in sintering the workpieces. The oven is provided with an argon purge.

Each test specimen, or workpiece, was prepared by cold-pressing boron carbide powder into a right circular cylinder measuring 1.0 cm in diameter by 1.0 cm high and having a density of about 50% of the theoretical density. After cold-pressing at 10,000 psi (68,900 kPa), each specimen was isostatically pressed at 50,000 psi (344,500 kPa), resulting in pellets which were slightly smaller than about 1 cm in diameter by 1 cm high. Each pellet then had a density of about 55% of theoretical density.

During heating, each workpiece was surrounded by zirconia insulation board, which served the dual purpose of containing the heat and preventing arcing between the workpiece and the wall of the microwave cavity. The external dimensions of the zirconia container, or sample holder, were 2.5 inches square (6.35 cm) by 4.5 inches (11.43 cm) high. The sample cavity inside the sample holder was about 1.25 cm in diameter by about 3.5 cm high. A 1/8 inch (0.32 cm) diameter hole in the sample holder allowed temperature measurements to be taken using the infrared optical sensor.

Each workpiece rested on a Carbocell® carbon disk, which rested on the zirconia of the sample holder. Also, a Carbocell® disk was placed on the top of the sample. The disks were about 1 cm in diameter and about 1 cm high. The Carbocell® disks served to reflect and concentrate microwave radiation in the sample. They also acted as oxygen getters to prevent oxidation since the argon purge of the oven cavity is not perfect. The bottom disk also prevented the contamination of the boron carbide by the zirconia insulation.

After pressing, each workpiece was heated from room temperature to peak temperature and held at about that temperature, using a reduced power level, for about 5 to 10 minutes before the microwave oven power was turned off and the sample allowed to cool to room temperature. The results are shown in the Table, which shows the greatest, or peak, temperature and the density achieved upon heating the test specimen to that temperature, for each of six specimens. Peak temperatures are in degrees Celsius and densities of the specimens are in percent of theoretical density.

TABLE

Temperature	Density
1840	59
1880	61
1950	82
1951	87
1983	93
2000	95

Since the two-color pyrometer used in the experimentation has a maximum temperature capability of 2,000°C, it is believed that the peak temperature of the 95% density sample was slightly greater than 2,000°C. Heating time varied with peak temperature and the power level at which the oven was operated. For example, a workpiece was heated to 2,000°C in about 4 1/2 minutes at a reduced power setting. The longest processing time for any of the specimens, consisting of heating time plus holding time, was 12 minutes. The volume of each specimen decreased by about 40% as a result of sintering.

After sintering, specimens were etched by an electrolytic technique which consisted of immersion in 1% potassium hydroxide solution for 10 to 15 seconds while a voltage of 15 to 30 V was applied. The etched specimens were observed with a microscope. The average grain size was about 20 microns. Several specimens were analyzed by x-ray diffraction and found to be pure boron carbide without any indication of phase separation.

Though all of the test specimens produced in the experimentation described herein were uniaxially pressed at about 10,000 psi (68,900 kPa) and then isostatically pressed at about 50,000 psi (344,500 kPa), it is expected that workpieces can be produced using pressures varying from about 5,000 to about 70,000 psi (34,500 - 482,300 kPa) and any method of pressing. It is expected that peak temperatures of from about 1,700 to about 2,200°C will be utilized in practice of the invention, with holding times varying from about 1 to about 30 minutes.

The energy requirement for microwave sintering to 95% density was about 0.906 kW-hours per gram of boron carbide. A sample of boron carbide which was hot-pressed (with inductive heating) used about 1.106 kW-hours per gram. Microwave sintering thus required 18% less energy, even though the hot-pressed sample was about 9 times larger than the microwave sintered specimen. Capitol costs for a microwave sintering facility are expected to be no more than about one-third the cost of a facility which utilizes conventional methods of production. Production rates for microwave sintering are expected to be much higher than those achieved with conventional processing since microwave sintering is about 10 to 50 times faster than inductive hot-pressing.

"Green," as in "a green shape" is a term used in the ceramic arts to indicate a partially finished object, such as an object which has been formed, but has not yet been densified.

The foregoing description of the invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed, since many modifications and variations are possible in light of the above teaching. It is intended that the scope of the invention be defined by the claims appended hereto.

ABSTRACT OF THE INVENTION

A method for forming boron carbide into a particular shape and densifying the green boron carbide shape. Boron carbide in powder form is pressed into a green shape and then sintered, using a microwave oven, to obtain a dense boron carbide body. Densities of greater than 95% of theoretical density have been obtained.