

POROUS SILICON CARBIDE CERAMICS USING FILLED POLYSILOXANES

This is a divisional of copending application Ser. No. 08/055,892 filed on May 3, 1993, now U.S. Pat. No. 5,283,019 issued Feb. 1, 1994.

BACKGROUND OF THE INVENTION

This invention relates to the preparation of porous ceramic bodies by the pyrolysis of curable organopolysiloxanes filled with silicon carbide powders and, optionally, organopolysiloxane curing agents and sintering aids. This process is advantageous in that both the green bodies and the resultant ceramics have high strengths.

Various methods for the production of porous silicon carbide ceramic bodies are known in the art. For instance, it is known to mold mixtures of large particle size silicon carbide and smaller size silicon carbide into a desired shape followed by pyrolyzing the molded mixture to form a ceramic. The particle size variation in this approach creates the desired pores. Similarly, it is known to mold a mixture comprising a carbonaceous binder, silicon carbide powder and a silaceous powder into the desired shape followed by pyrolysis. In this approach, the carbon formed by pyrolysis of the carbonaceous binder reacts with the silaceous binder to form silicon carbide, thus, leaving pores in the sinter. Neither of these approaches, however, encompasses the use of organopolysiloxanes as binders to form porous ceramics.

Suganuma et al. in the journal of *Materials Science*, 28 (1993) pp. 1175-1181, teach the formation of porous silicon carbide bodies by pyrolyzing a mixture of SiC powder, polysilastyrene and an organic binder. The reference, therefore, differs from the present invention in that it requires the use of an organic binder and it does not mention the use of polyorganosiloxanes.

Atwell et al. in U.S. Pat. No. 4,888,376 teach that high density silicon carbide bodies ($>2.4 \text{ g/cm}^3$) can be formed by molding a mixture comprising an organopolysiloxane, a sintering aid, an organopolysiloxane curing agent and silicon carbide powder and then heating the molded mixture to a temperature above about 1900°C . This reference, however, teaches that the bodies are highly densified compared to the high strength porous bodies described herein.

The present invention provides porous ceramic products using organopolysiloxanes as binders. Both the green bodies and the resultant ceramics have high strengths.

SUMMARY OF THE INVENTION

This invention relates to a method of preparing a porous silicon carbide ceramic body. The method comprises forming a mixture comprising silicon carbide powder, a preceramic organopolysiloxane, and, optionally, an organopolysiloxane curing agent and/or a sintering aid into a green body having the desired shape. The green body is then sintered in an inert atmosphere at a temperature sufficient to form a porous sintered body having a density less than 2.4 g/cm^3 and an open porosity greater than about 25%.

DETAILED DESCRIPTION OF THE INVENTION

The present invention is based on the unexpected discovery that mixtures of organopolysiloxanes and

silicon carbide powder can be used to prepare high strength green bodies which can be fired to form high strength, monolithic, porous ceramic bodies. This was particularly unexpected since the prior art does not teach the use of organopolysiloxanes in the formation of porous ceramics nor the formation of the high strength bodies claimed herein.

The sintered bodies produced from the practice of this invention have controlled open porosity (greater than about 25%), low firing shrinkage with low shrinkage variation, a unique pore and grain morphology, high modulus and high strength (often above 25 ksi (ksi=kpsi=1000 pounds per square inch)), and good oxidation and corrosion resistance. The density of these materials is less than about 75% of the theoretical density of silicon carbide (3.21 g/cm^3), i.e., densities less than about 2.4 g/cm^3 . As such, these materials are useful, for example, as high temperature filters, as supports for catalysts in chemical reactions and in structural applications where high strength porous bodies are required.

The high green strength obtained by the process of the present invention is a significant advantage which allows the green bodies to be handled and further processed and shaped prior to the final sintering or pyrolysis step. Generally, green strengths of 500 psi or more may be obtained in the practice of this invention. Additionally, the green body may be cured prior to sintering to obtain even stronger green bodies.

The mixtures used to prepare the porous bodies of the present invention comprise an organopolysiloxane filled with silicon carbide powder and, optionally, an organopolysiloxane curing agent and/or a sintering aid. According to the invention, this mixture is molded into the desired shape and pyrolyzed. To prevent densification during the pyrolysis and, thus, provide a porous body, the present inventors have discovered that it is necessary to control either the composition of the mixture or the processing conditions. This can include selecting an organopolysiloxane which produces limited amounts of free carbon, limiting the amount of sintering aid and/or limiting the pyrolysis temperature, each of which is described hereinafter. By taking these steps, the desired porous bodies are formed.

The organopolysiloxanes useful herein are generally well known in the art. The primary requirement of such polymers is their capability of being converted to ceramic materials with a ceramic char yield greater than about 20 weight percent (the ceramic char is that material remaining after pyrolysis of the organopolysiloxane to 1800°C . and the ceramic char yield is the weight percent of ceramic char compared with the weight of organopolysiloxane pyrolyzed). However, since there is less shrinkage with higher char yield polymers, it is preferred that organopolysiloxanes having char yields greater than about 40 weight percent be employed.

The organopolysiloxanes used herein should also generally yield a ceramic char having at least a stoichiometric amount of silicon and carbon. Organopolysiloxanes which produce silicon rich chars (i.e., greater than a stoichiometric amount of silicon) are generally not useful as they result in ceramic bodies having lower strengths and poor oxidation resistance. Organopolysiloxanes which produce carbon rich chars (i.e., greater than a stoichiometric amount of carbon) may be used herein. However, if the amount of free carbon (i.e., that amount of carbon above the stoichiometric amount in SiC) produced by such polymers is greater than about