

## LOW TEMPERATURE JOINING OF CERAMIC COMPOSITES

This is a division of Ser. No. 08/631,259, filed Apr. 12, 1996, now U.S. Pat. No. 5,858,144.

### CONTRACTURAL ORIGIN OF THE INVENTION

The United States Government has rights in this invention pursuant to Contract No. W-7405-ENG-82 between the U.S. Department of Energy and Iowa State University, Ames, Iowa, which contract grants to the Iowa State University Research Foundation, Inc. the right to apply for this patent.

### FIELD OF THE INVENTION

The present invention relates to joining of ceramics and ceramic composites at relatively low temperatures and in ambient air atmosphere, if desired, and to bonding agents to this end.

### BACKGROUND OF THE INVENTION

In recent years, there has been great commercial interest in and government support for the use of ceramic composites, particularly SiC continuous fiber ceramic composites (CFCC) materials, for structural applications in high temperature and/or corrosive environments. This materials substitution trend recognizes that traditional stainless steels and superalloys have reached their operational limits and that further enhancements of process efficiency in applications such as power generation heat exchangers requires the high temperature and chemical stability and tolerance of SiC CFCC materials. Unfortunately, the lack of convenient joining methods for SiC CFCC materials has limited their service applications because of the difficulty of winding large, complex SiC fiber preforms and of infiltrating the preforms with SiC matrix material.

A goal of the art has been to assemble complex structures from a set of simple SiC CFCC component shapes joined together so as to collectively provide the desired complex structural shape with sufficient strength and toughness to be tolerant of severe environments and rough handling, enabling the potential of SiC CFCC materials to be more fully realized.

To-date, joining of monolithic SiC shapes has been accomplished by a variety of techniques including direct diffusion bonding, co-densification of interlayer and green bodies, diffusion welding or brazing with boride, carbide, and silicide interlayers, hot pressing of sinterable SiC powder, bonding with polymeric precursors, brazing with oxide, reactive metal bonding, and active metal brazing. These joining techniques typically require prolonged exposure to high temperatures above 1600 degrees C. and/or strict inert joining atmospheres and many require a bonding agent including a solvent to provide proper bonding material viscosity. Existing commercial SiC CFCC materials almost universally employ "NICALON" SiC fibers which contain some residual SiO<sub>2</sub> and free C.

Many investigators have reported that these residual phases contribute to a fiber degradation reaction that has an onset temperature of about 1200 degree C. Thus, any SiC CFCC joining technique that requires firing temperatures above 1200 degrees C. is not desirable because some degree of fiber degradation and a resulting decrease in strength of the CFCC may occur.

An object of the present invention is to provide a method of joining ceramic and ceramic composite materials at

relatively low temperatures that avoids degradation of the ceramic material; e.g. not exceeding 1200 degrees C. for joining SiC fiber CFCC materials.

Another object of the present invention is to provide a method of joining ceramic and ceramic composite materials that can be practiced in ambient air, if desired, in order to facilitate fabrication of structures on-site under a wide variety of field assembly conditions.

Still another object of the present invention is to provide a bonding agent for joining or repairing ceramic and ceramic composite materials that is effective to promote mechanical bond toughness and elevated temperature strength and that can be used at relatively low temperatures without the need for infiltration and without the need for solvents.

Still a further object of the present invention is to provide a bonded structure characterized by a joint microstructure having improved strength and toughness by virtue of including relatively soft, compliant phase regions dispersed in a ceramic joint matrix.

### SUMMARY OF THE INVENTION

The present invention provides a method of joining similar or dissimilar ceramic and ceramic composite materials at relatively low joining temperatures using a solventless, three component bonding agent effective to promote mechanical bond toughness and elevated temperature strength to operating temperatures of approximately 1200 degrees C.

A bonding agent in accordance with an embodiment of the present invention comprises a preceramic precursor, an aluminum bearing powder, such as aluminum metal powder, and boron powder in selected proportions to provide desired mechanical bond toughness and elevated temperature strength for a given service application.

In accordance with a method embodiment of the present invention, the bonding agent is disposed as an interlayer between similar or dissimilar ceramic or ceramic composite materials to be joined and the interface is heated to a temperature not exceeding about 1200 degrees C. to pyrolyze the preceramic polymer so as to form a strong and tough bond joint between the materials.

The method advantageously can be practiced in ambient air atmosphere using an open flame of a propane or other torch to heat the materials to the bonding temperature. This facilitates fabrication of structures on-site under a wide variety of field assembly conditions. Alternately, an inert or air atmosphere can be used in a firing furnace.

The bond joint produced in accordance with another embodiment of the present invention is characterized by a composite joint microstructure having relatively soft, compliant aluminum bearing phase regions, such as aluminum-silicon alloy particles, disposed in a ceramic matrix, such as SiC, and exhibiting substantially improved joint strength and toughness.

In accordance with another method embodiment of the present invention, the bonding agent is used as a filler material to repair a crack, pinhole or other depression in a ceramic and ceramic composite member and heated to a temperature not exceeding about 1200 degrees C. to form a strong and tough repair filler bond with the ceramic material.

The present invention is especially useful for, although not limited to, bonding of SiC continuous fiber ceramic composite (CFCC) materials together without harmful degradation of the SiC fibers so as to form a desired structure having sufficient strength and toughness that is tolerant of severe environments and handling and service stresses. The