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CRADA Final Report
for
CRADA Number ORNL94-0250

GELCASTING OF CRYSTAR® SILICON CARBIDE CERAMICS

S. D. Nunn
Oak Ridge National Laboratory

C. A. Willkins
Saint-Gobain/Norton Industrial Ceramics Corporation

Prepared by the
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831
managed by
Lockheed Martin Energy Research Corporation
for the
U.S. Department of Energy
under contract DE-AC05-96OR22464

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ORNL-27 (3-96)

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Research sponsored by the Advanced Automotive Materials Program,
DOE Office of Transportation Technologies, under contract DE-AC05-96OR22464 with
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ABSTRACT

This Cooperative Research and Development Agreement (CRADA) was undertaken to assess the applicability the gelcasting process for forming ceramic green bodies using Saint-Gobain/Norton Industrial Ceramics Corporation's proprietary CRYSTAR® silicon carbide powder. A gelcasting process, specifically tailored to Saint-Gobain/Norton's powder composition, was developed and used successfully to form green bodies for property evaluation. This preliminary evaluation showed that the gelcast material had characteristics and properties comparable to Norton's baseline material. Wafer carrier molds were received from Norton for gelcasting a complex-shaped configuration with CRYSTAR® silicon carbide. Gelcasting experiments showed that Norton's standard plaster of paris molds were incompatible with the gelcasting process. Mold surface treatments and the use of alternative castable mold materials were investigated, however, a successful process was not identified. The highest quality parts were cast in either glass or aluminum molds.

OBJECTIVES

The desired result of this CRADA was the successful demonstration of the gelcasting process for forming ceramic green bodies using the Participant's powder composition. Success was be judged by the quality of the castings and by the measurement of the properties of the green and fired gelcast samples. The Participant also gained valuable hands-on experience in using the gelcasting process which will aid in transitioning the technology to production at the Participant's facilities.

Meeting Objectives

The majority of the technical objectives of this CRADA were met in a timely manner with good interaction between ORNL and Saint-Gobain/Norton.

CRADA Benefit to DOE

Conduct of research under this agreement served to increase our understanding of the important ceramic powder characteristics and processing parameters which govern successful application of the gelcasting method.

TECHNICAL DISCUSSION

Introduction

The purpose of this Cooperative Research and Development Agreement (CRADA) is to assess the applicability the gelcasting process for forming ceramic green bodies using Saint-Gobain/Norton Industrial Ceramics Corporation's proprietary CRYSTAR® silicon carbide powder. The gelcasting process has been used successfully to form ceramic green bodies from numerous ceramic materials, including silicon nitride, silicon carbide, alumina, zirconia, etc. During these prior investigations, it has been observed that the gelcasting process needs to be tailored to each specific material. This is primarily due to the fact that gelcasting is a chemical process and any variation in the chemistry of the system will affect the characteristics of the process.

In order to gain confidence in the applicability of gelcasting using Saint-Gobain/Norton's proprietary ceramic composition and processing, and to minimize the learning curve required for tailoring the process to their material, a cooperative research effort was proposed. ORNL provided the expertise and equipment for working with the gelcasting process. Saint-Gobain/Norton provided ceramic materials, specialty molds, and final processing and testing expertise to evaluate the gelcast material. The desired result of this CRADA was the successful demonstration of the gelcasting process for forming ceramic green bodies using Saint-Gobain/Norton's CRYSTAR® silicon carbide powder composition.

Gelcasting Process Development

The silicon carbide powder to be used for this project was received from Norton at ORNL along with a description of Norton's standard processing procedure for preparing a slurry from the powder. Two trial gelcasting batches were prepared. The standard Norton procedure for preparing the silicon carbide powder slurry was used to produce a 72 vol. % solids-loaded slip. The gel-forming chemicals were then added to the batch in preparation for casting. Two gelcasting formulations were evaluated: Methacrylamide monomer with methylene bisacrylamide crosslinker in a 6:1 ratio (MAM/MBAM) and MAM with polyethylene glycol dimethacrylate crosslinker in a 3:1 ratio (MAM/PEG). Both formulations produced low-viscosity slurries which were easily de-aired and cast. Simple disks and flat plate specimens were cast and gelled successfully. The gelled bodies had sufficient strength to retain the cast shape without deforming during unmolding and drying. The green density of the cast parts after drying was 72% of the theoretical density indicating virtually no shrinkage during the gelling and drying steps.

Gelcast CRYSTAR® silicon carbide plates were prepared for preliminary property evaluation. As in the previous trials with this material, the standard Norton procedure for preparing the silicon carbide powder slurry was used. Plates were fabricated using both the MAM/MBAM and the MAM/PEG gel-forming chemical systems. The gelcast plates were dried and then shipped to Norton for firing and evaluation.

Preliminary Property Determination

Four flat plates which were gelcast from the CRYSTAR® silicon carbide powder were given an initial green body evaluation followed by the first firing step. They then underwent a siliconizing step to produce the finished material for characterization and evaluation. The evaluation included tests of the density, fracture strength, and fracture toughness of the fired samples. The test results, as well as dimensional and shrinkage data, compared favorably with the standard material.

This completed the first phase of the project and a decision was made to proceed to the second phase which entailed the fabrication of complex shapes.

Complex Part Fabrication

Craig Willkens, the Norton principal investigator, visited ORNL to obtain hands-on experience with the gelcasting process. A number of simple shapes were cast and gelled. Mold materials and procedures for casting complex-shaped silicon wafer carriers were discussed.

A wafer carrier mold was received from Norton for gelcasting a complex-shaped configuration with CRYSTAR® silicon carbide. The mold was made of plaster of paris and needed to be surface-sealed before it could be used in the gelcasting process. A number of sealers were evaluated, and an aerosol acrylic spray was found to be most effective. A gelcasting slurry was prepared and cast in the mold. The mold cavity was filled simply by pouring the slurry into the mold. Gelling was complete after approximately 30 min. at 65°C. Two problems were encountered: First, a small portion of the slurry flowed into the parting seams of the mold after the mold was placed in the oven to gel. This resulted in an incompletely filled mold and corresponding defects in the cast part. Improved mold sealing or provide a reservoir of slurry were needed to prevent these flaws in later castings. The second problem was related to removing the cast part from the mold. The casting was cracked while trying to pry the part off of the mold surface. It was later found that if the casting was left on the mold and allowed to begin drying under ambient conditions that the part freed itself from the mold surface, thus eliminating the damage.

A new plaster of paris wafer carrier mold was received from Norton. Once again, the mold surface was sealed with an aerosol acrylic spray. A gelcasting slurry containing 68 vol. % solids was prepared for the first casting trial with the new mold. The reduced solids content lowered the viscosity of the slurry and allowed easier pouring of the slurry into the mold. The mold was filled with the slurry and the gelling was complete after approximately 30 min. at 65°C. Removal of the part from the mold was difficult due to part adhesion to the mold surfaces.

An additional mold release agent was used for the second casting trial with the new wafer carrier mold. Although the mold was coated liberally with the release agent, the cast sample adhered to the mold surface as in the previous trial. The sample could not be removed from the mold without damaging the casting. After discussion of the problem with Norton, it was decided that they would provide simple tile casting molds made from different plaster of paris formulations to test for ease of mold release. Once an acceptable formulation was identified, a new wafer carrier mold could be fabricated for continuation of the gelcasting trials.

Trial runs were conducted to evaluate the mold release characteristics of several variations of plaster of paris molds. Four different mold compositions were received from Norton containing different percentages of an additive in the plaster of paris to modify the pore structure. These molds were surface-sealed with either an aerosol acrylic lacquer or an aerosol polyurethane coating to prevent absorption of the water from the gelcasting slurry. The sealed molds were tested either with or without a mold release agent. These variations resulted in sixteen different combinations for evaluation. A gelcasting slurry of CRYSTAR® silicon carbide was cast in the molds and gelled. After cooling, the cast parts were removed from the molds. The results indicate that the pure plaster of paris composition, sealed with polyurethane, and coated with the mold release agent provided the best release characteristics. A new wafer carrier mold of the pure plaster of paris composition was fabricated by Norton and was shipped to ORNL for continuation of the gelcasting study.

The new plaster of paris wafer carrier mold which was received from Norton was surface-sealed with the polyurethane coating spray. After the coating had dried, mold release was applied to the surface and the mold was assembled for casting. A slurry of the CRYSTAR® silicon carbide was prepared using the MAM/MBAM gelcasting chemical system. This slurry was then cast in the mold and gelled at 65°C. After cooling, the mold was disassembled. As with the previous casting trials, the gelled part adhered to the surface of the mold and could not be removed without damaging the casting. Attempts to improve casting release by increasing the amount of mold release agent were only partially successful. Applying sufficient mold release so the castings were more easily removed from the mold resulted in a poor surface finish on the cast parts.

As an alternative approach for improving casting quality, a number of new mold variations were evaluated. These included coating the plaster of paris molds with a wax layer; using a castable, aluminum-filled polyurethane mold; and using a silicone rubber mold. Initially, all of these variations showed a thin liquid layer at the surface of the gelcast parts. Experiments showed that this liquid layer could be significantly reduced by increasing the polymerization initiator content in the gelcasting slurry. A number of castings were removed successfully from the molds, but some surface imperfections were observed. Additional improvement would be required to produce high quality castings. Trial castings using simple mold configurations showed that excellent release and surface finish could be obtained when using either a glass or an aluminum mold. Norton will consider the possibility of procuring an aluminum mold for the wafer boat to use in future gelcasting trials.

Conclusions

This Cooperative Research and Development Agreement (CRADA) was undertaken to assess the applicability the gelcasting process for forming ceramic green bodies using Saint-Gobain/Norton Industrial Ceramics Corporation's proprietary CRYSTAR® silicon carbide powder. A gelcasting process, specifically tailored to Saint-Gobain/Norton's powder composition, was developed and used successfully to form green bodies for property evaluation. This preliminary evaluation showed that the gelcast material had characteristics and properties comparable to Norton's baseline material. Wafer carrier molds were received from Norton for gelcasting a complex-shaped configuration with CRYSTAR® silicon carbide. Gelcasting experiments showed that Norton's standard plaster of paris molds were incompatible with the gelcasting process. Mold surface treatments and the use of alternative castable mold materials were investigated, however, a successful mold material was not identified. The highest quality parts were cast in either glass or aluminum molds.

Report of Inventions

There were no inventions developed under this agreement.

Commercialization Possibilities

The gelcasting process developed under this agreement may be commercialized at some time in the future, however, there are no such plans currently in place.

Plans for Future Collaboration

No plans have been made for future collaboration.

CONCLUSIONS

The majority of the technical objectives of this CRADA were met in a timely manner with good interaction between ORNL and Saint-Gobain/Norton. A gelcasting process, specifically tailored to Saint-Gobain/Norton's powder composition, was developed and used successfully to form green bodies for property evaluation. This preliminary evaluation showed that the gelcast material had characteristics and properties comparable to Norton's baseline material. Wafer carrier molds were received from Norton for gelcasting a complex-shaped configuration with CRYSTAR® silicon carbide. Gelcasting experiments showed that Norton's standard plaster of paris molds were incompatible with the gelcasting process. Mold surface treatments and the use of alternative castable mold materials were investigated, however, a successful mold material was not identified. The highest quality parts were cast in either glass or aluminum molds.

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