

ANL/ET/CA.- 91636

CONF-970111--10

MAGNETIC RESONANCE IMAGING OF GEL-CAST CERAMIC COMPOSITES*

by

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Proceedings of the 21st Annual Cocoa Beach Conference and Exposition on Composites, Advanced Ceramics, Materials, and Structures, Cocoa Beach, Florida, Jan. 12-16, 1997, to be published in the Ceramic Engineering and Science Proceedings.

*We are pleased to acknowledge that the financial support for this work was provided by the U.S. Department of Energy, Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Transportation Systems, as part of the Ceramic Technology Project of the Advanced Materials Development Program, under Contract W-31-109-ENG-38.

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ABSTRACT

Magnetic resonance imaging (MRI) techniques are being employed to aid in the development of advanced near-net-shape gel-cast ceramic composites. MRI is a unique nondestructive evaluation tool that provides information on both the chemical and physical properties of materials. In this effort, MRI imaging was performed to monitor the drying of porous green-state alumina - methacrylamide-N,N'-methylene bisacrylamide (MAM-MBAM) polymerized composite specimens. Studies were performed on several specimens as a function of humidity and time. The mass and shrinkage of the specimens were also monitored and correlated with the water content.

INTRODUCTION

The application of the generic gel-cast ceramic process is of considerable interest for the high-volume manufacturing of near-net-

shape complex components.[1,2] The technique can be employed for a diverse group of materials to produce both monolithic and composite ceramic components. In this processing method, a ceramic powder is mixed with an organic monomer in solution. An initiator is then added to begin the polymerization reaction and cast into a mold. Upon solvent removal the components exhibit minimal shrinkage, high structural integrity, and can be readily machined using conventional tooling.

Magnetic resonance imaging is a 3-D tomographic nondestructive evaluation technology that is capable of (1) mapping ceramic/polymer homogeneity, (2) real-time imaging of the polymerization process, (3) mapping of voids and flaws, and (4) measuring various physical properties (e.g., moisture, size, degree of polymerization, viscosity, density, and specimen strength). MRI techniques have previously provided information on both the physical and chemical properties of a diverse number of materials including wood, polymers, porous structures, and ceramics.[3,4] Studies in the ceramics area have included efforts in both injection

molding and slip-cast materials.[5,6] Recently, in-situ monitoring of the polymerization of methacrylamide gels has been demonstrated.[7]

In this program MRI is applied to the methacrylamide polymer-ceramic composite system. The specific tasks in this program include investigating the phenomena of green body drying as a function of time and humidity as well as the effects of component compositions on the physical strength of the green body ceramic. Drying studies of other gel-cast ceramics have previously been performed.[8] This study measured the drying rate of the sample by mass measurements. In contrast, our study employs MRI to monitor the water migration through the sample by selecting slices through the specimen. The goal of this program is to aid in the development and optimization of large-scale gel-cast processing techniques.

EXPERIMENTAL



Figure 1. Photographs depicting ANL's anodized aluminum breakaway mold (left), casting of the gel- Al_2O_3 matrix (middle), the resultant cast green-state gel-cast ceramic composite specimen (right).

The polymer premix solution consisted of a 15% (wt) methacrylamide-N,N'-methylene bisacrylamide (MAM-MBAM) (6:1 wt MAM-MBAM ratio) polymer system. The slurry mixture was 50% (vol) alumina (RC-HP-DBM, Reynolds), and 50% (vol) of the premix polymer solution. A high-shear blender and the dispersant Darvan C (R. T. Vanderbilt Co. Inc.) were employed to facilitate the suspension of the alumina into the polymer solution. The resultant slurry was degassed in a vacuum dessicator and then the initiator (ammonium persulfate) and accelerator

(tetramethylethylenediamine) agents were added to the slurry to begin the polymerization reaction. The slurry was immediately poured into molds. The molds consisted of 1" high x 2" diameter cylinders of anodized aluminum which were treated with a mold release agent (Epoxy Parfilm Ultra II, Price Driscoll Corp.). All image specimens were prepared at ambient temperature in a nitrogen environment. The specimens were dried slowly in constant humidity chambers that were prepared using standard saturated salt solution techniques. [9]

The magnetic resonance imaging experiments were performed with a commercial spectrometer (DMX-300, Bruker Instr. Inc.) coupled to a 2 Tesla horizontal imaging magnet system. The data were acquired by employing slice-selected gradient-recalled imaging sequences.

Vials containing various H₂O-D₂O solutions were included in the image to provide intensity standards for proton calibration. Image intensities were determined from a circular region-of-interest which covered approximately one half of the interior portion of the specimen image. Imaging experiments were repeated daily.

RESULTS

MRI imaging results obtained on three separate gel-cast ceramic composites are presented in Figure 2. The images represent a drying time span of ten days. The variations of gray scale intensity observed throughout the images represent differences in water content of the green body. Lighter regions indicate high water concentrations while dark areas indicate the absence of protons. Inhomogeneities of the intensity in the images are possibly the result of voids or bubbles in the cast specimens, or nonuniform dispersion of ceramic powders in the polymer matrix. The trend of intensity decay as a function of drying time for each relative humidity can be observed from the curves in Figure 3A. The image intensity observed in the lower humidity specimen decayed at a faster rate than in higher humidities. Additionally, this curve displayed a much larger second order component than in the higher humidities. Furthermore, the normalized masses of the specimens are observed to follow the same trends, see Figure 3B. Therefore, the gel-cast specimens

dried faster in lower humidities. The specimen diameters are shown in Figure 3C. The majority of the shrinkage of the components occurs during the initial 5% of the specimen weight loss. In this work we have shown that MRI can successfully image gel-cast ceramic composite systems to yield information about the chemical and physical properties of the sample. Future efforts will be devoted to additional drying studies on samples with more complex shapes and further investigation of the polymerization process in the gel-cast ceramic component. This will include measurements of other NMR parameters as well as a comparison of drying trends with conventional ceramic forming processes.

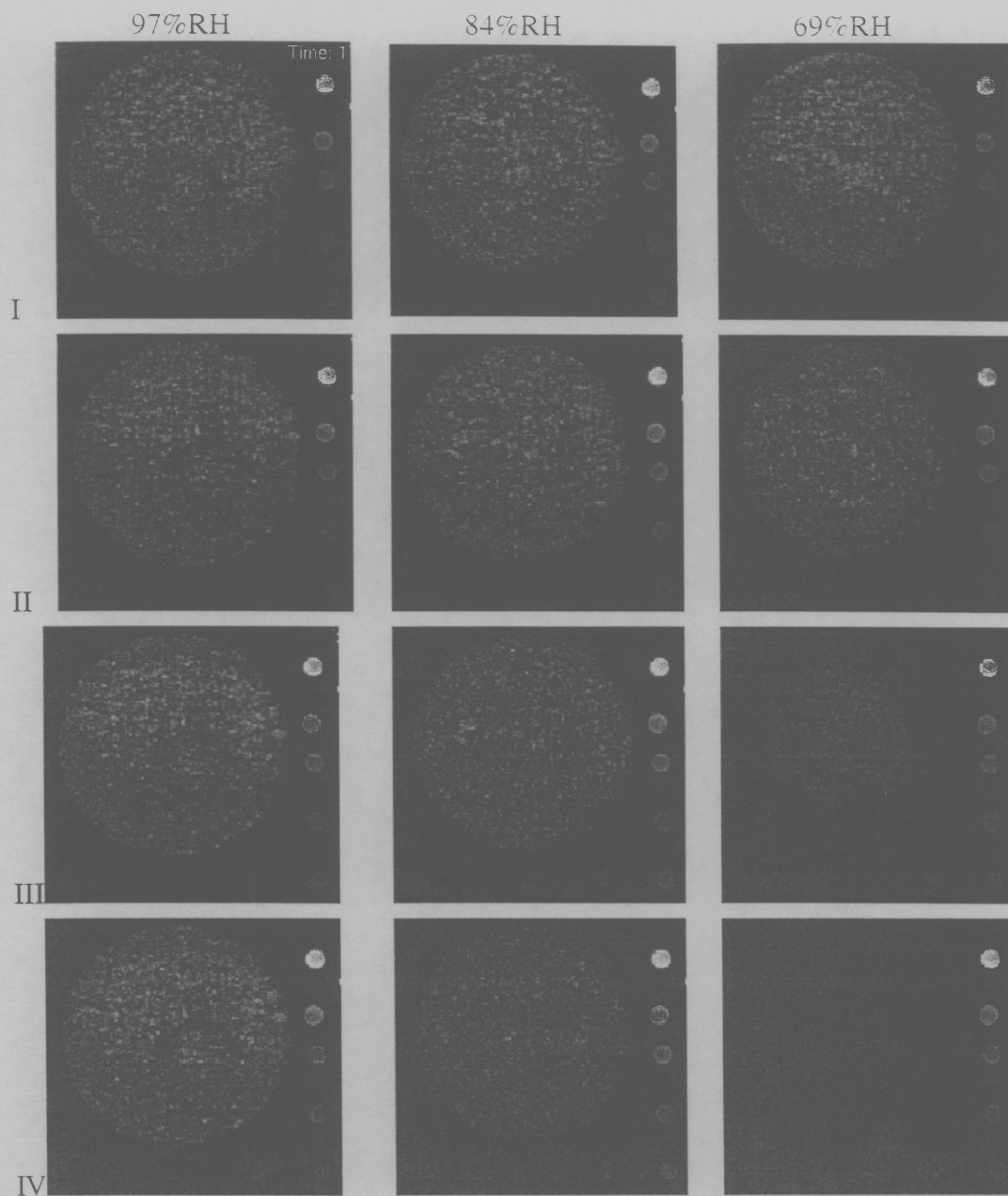
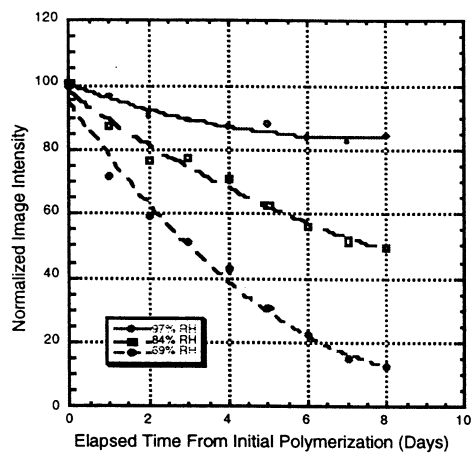
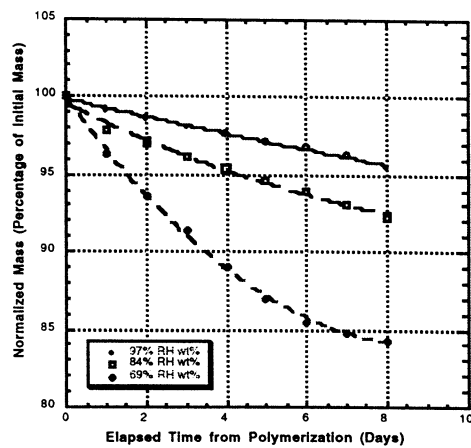


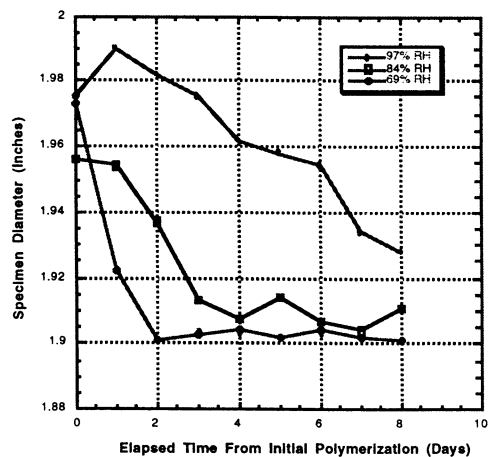
Figure 2. Magnetic resonance images acquired from a 5 millimeters slice selection of three separate specimens as a function of time and humidity. Columns I through III represent specimens dried at 97, 84, and 69% relative humidities, and rows I through IV represent the images obtained at 2, 4, 6, and 8 days respectively. Vials containing water concentrations of 55, 40, 30, 25, 20, and 10% water are shown descending vertically to the right of the images.



A



B



C

Figure 3. Plots of MRI image intensity (A), specimen mass (B) and sample diameter (C) as a function of relative humidity and time.

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ACKNOWLEDGMENTS

We wish to thank Dr. Ogbemi O. Ometete and Ms. Claudia A. Walls of Oak Ridge National Laboratory , Oak Ridge TN, for their support and recommendations throughout this research program. We are also pleased to acknowledge that the financial support for this work was provided by the U.S. Department of Energy, Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Transportation Systems, as part of the Ceramic Technology Project of the Advanced Materials Development Program, under Contract W-31-109-ENG-38.