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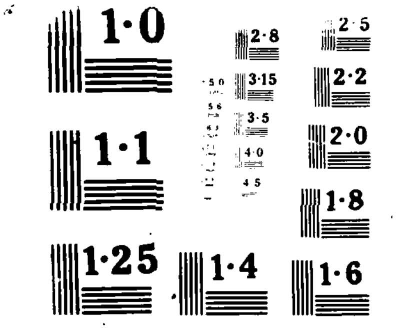

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Hot Isostatic Pressing, alumina, pore stability, dihedral angle, grain boundary diffusion, volume diffusion, microstructure heterogeneities, final stage densification, liquid phase sintering, magnesium-aluminosilicate glass, solution-precipitation, gas diffusion, liquid phase redistribution.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
The final stage densification of undoped Al_2O_3, MgO-doped Al_2O_3 and magnesium aluminosilicate glass/Al_2O_3 composites by hot isostatic pressing unencapsulated, closed porosity samples has been investigated. The conditions for pore stability have been reviewed for these systems that are representative of ceramics densifying by solid state diffusion and solution-precipitation mechanisms. An extensive series of experiments were conducted and the results used to test the validity of models derived to understand the effect of (1) dihedral angle, effective stress, grain size distribution and pore/grain size ratio on pore stability during hot

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isostatic pressing of pure and MgO-doped alumina.

Models explained the influence of these parameters although 100% was not observed as a result of porous heterogeneities exceeding the critical pore/grain size ratio of density ~2.3.

(2) Dihedral angle, effective stress, grain size, volume percent liquid, gas diffusion and liquid redistribution during hot isostatic pressing of alumina glass composites.

Models for pressureless and pressure enhanced solution precipitation were derived to incorporate the effective stress. Experiments at various pressures agree well with the new models and demonstrate that densification is limited by the interface reaction for short (i.e., <1 hr) HIP times. Longer HIPing experiments resulted in dedensification as a result of pressure enhanced gas diffusion into the glass matrix. Models were derived to predict the effect of gas diffusion on densification. At >98% relative density macropore (100 μm diameter) closure can occur by pressure driven liquid redistribution. Depending on matrix grain size the macropores can be filled completely by liquid or a liquid/particle mixture.

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HOT ISOSTATIC PRESSING OF SINTERED CERAMICS

FINAL TECHNICAL REPORT

Gary L. Messing, Kevin G. Ewsuk and Oh-Hun Kwon

May 10, 1986

U.S. ARMY RESEARCH OFFICE

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The Pennsylvania State University

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Hot Isostatic Pressing of Sintered Ceramics

This final report is presented in two separate, but related, sections. The two sections correspond to the two PhD dissertations that were completed during this research contract. The two dissertations summarized herein are as follows:

Kevin G. Ewsuk, "Final Stage Densification of Alumina During Hot Isostatic Pressing," Ph.D. in Ceramic Science, May 1986.

Oh Hun Kwon, "Liquid Phase Sintering and Hot Isostatic Pressing of an Alumina-Glass Composite: Modeling and Experiment," Ph.D. in Ceramic Science, August 1986.

Publications that have been published or submitted for publication include:

O.-H. Kwon and G. L. Messing, "Microstructural Changes in Hot Isostatically Pressed Alumina-Glass Composites," J. Am. Ceram. Soc. 67(3) C43-45 (1984)

O.-H. Kwon and G. L. Messing, "HIP of Liquid Phase Sintered Ceramic Composites," in Tailoring Multiphase and Composite Ceramics eds. G. L. Messing, R. F. Tressler, C. G. Pantano and R. E. Newnham, Plenum Press, New York (1986).

O.-H. Kwon and G. L. Messing, "Macropore Filling During Hot Isostatic Pressing of Liquid Phase Sintered Ceramics," in Sintering-Theory and Practice-II eds. H. Palmour and G. Kuczynski, Plenum Press, New York (1986).

At least 9 more manuscripts based on these two dissertations will be submitted for publication by the end of 1986 and include:

K. G. Ewsuk and G. L. Messing, "Theoretical Predictions of Pressure Enhanced Final Stage Densification"

K. G. Ewsuk and G. L. Messing, "Pore Shrinkage in Alumina During Hot Isostatic Pressing"

K. G. Ewsuk and G. L. Messing, "Final Stage Densification of MgO-doped Alumina by Hot Isostatic Pressing"

O.-H. Kwon and G. L. Messing, "Models for Final Stage Densification by Liquid Phase Sintering"

O.-H. Kwon and G. L. Messing, "Liquid Phase Sintering of Alumina-Glass Composites"

O.-H. Kwon and G. L. Messing, "Models for Pressure Enhanced Final Stage Densification of Alumina-Glass Composites"

O.-H. Kwon and G. L. Messing, "HIP Densification of Liquid Phase Sintered Alumina-Glass Composites"

O.-H. Kwon and G. L. Messing, "Liquid Phase Redistribution in Ceramics by Hot Isostatic Pressing"

O.-H. Kwon and G. L. Messing, "Gas Diffusion and Dissolution Effects on HIP Densification of Liquid Phase Sintered Ceramics"

Part I - Final Stage Densification of Alumina During Hot Isostatic Pressing

Introduction

Although it has long been recognized that a particulate mass subjected to elevated temperature for an extended period of time will densify to form a cohesive structure, only recently have efforts been made to understand this process. Over the past three decades, substantial progress has been made in both developing and applying conventional sintering theory. Consequently, there is now a more comprehensive understanding of how and why densification occurs. However, while conventional sintering theory has developed considerably, the theoretical aspects of sintering with pressure have largely been ignored.

It is well known that pressure sintering techniques, such as hot pressing (HP) and hot isostatic pressing (HIPing), can be used to produce dense, fine-grained microstructures possessing properties superior to those obtained by conventional sintering. The potential benefits of processing with pressure in conjunction with an increased desire to control microstructure and optimize properties during processing has generated a growing interest in HIP processing in particular. Unfortunately, due to a limited understanding of pressure sintering theory, the full potential of HIP processing has yet to be realized.

To develop a more comprehensive understanding of pressure sintering, the objectives of this study were:

(1) To establish a suitable model to describe final stage densification during HIPing, and subsequently analyze this model with respect to material properties and processing variables to ascertain the critical densification parameters.

(2) To determine the applicability of the pressure sintering model for predicting final stage densification in real systems during HIPing.

Summary

Densification theory is reviewed, and a model for predicting final stage densification during hot isostatic pressing (HIPing) is presented. An analysis of the model shows temperature, pressure, and grain size to be the critical processing parameters with respect to densification during HIPing. Results from experiments conducted on undoped and 0.40 wt% MgO-doped alumina are used to test the applicability of the model for predicting final stage densification in real systems during HIPing.

Sintered, MgO-doped alumina with a median grain size of 1.1 um densifies to 99.4% theoretical density within 7.5 to 15 minutes of HIPing at 1550oC and 69 MPa. In comparison, the model predicts that ~6 minutes is required to achieve the same level of densification for these conditions. Comparable agreement is also observed between model predictions and the results obtained from HIP experiments conducted at 1400 and 1450oC and 69 MPa, and at 1600oC and 124 MPa. Differences between the predicted and observed densification kinetics during HIPing are explained on the basis of the microstructural heterogeneities present in the systems studied, including surface energy anisotropy, particle packing defects, processing defects and grain and pore size distributions. Minimizing the influence of these microstructural heterogeneities on densification results in better agreement between experiment and theory, indicating that the proposed model yields an accurate description of final stage densification in an ideal system during HIPing.

Densification during HIPing is shown to occur rapidly, primarily via the preferential elimination of fine porosity. Due to the annihilation of larger pores than can be eliminated by sintering without pressure, higher densities are obtained. The inability to eliminate gross porosity and other energetically stable pores during HIPing results in less than theoretical end point densities. Theoretical arguments are presented which demonstrate that equiaxed pores larger than ~2.3 times the grain size cannot be eliminated during HIPing.

The applied HIP pressure is shown to enhance the rate of densification, and improve the ultimate density obtained after sintering. Additionally, the increased driving force for densification associated with the application of pressure during sintering promotes densification at lower temperatures and without grain growth.

Part II - Liquid Phase Sintering and Hot Isostatic Pressing of an Aluina-Glass Composite: Modeling and Experiments

Introduction

Powder compacts of high temperature materials become useful monoliths after they are subjected to a high temperature for densification. While this process, sintering, is one of the oldest technologies, scientific study of sintering has been limited to the last half century. Until now, a large amount of work has been done both in theory and in experiment to understand the process more quantitatively. Recent demand for high performance technical materials, has resulted in significant progress in this technology from powder synthesis to the tailoring of the final microstructure. For designing and manufacturing of defect-free high technology structural materials and molecular engineering materials, establishment of material preparation technology is increasingly important. Relationships between critical processing parameters as well as related material properties should be known comprehensively to meet the demand.

Materials for high performance application are either single phase or multiphase depending on application. For designing and practical purposes, multiphase (composite) materials have been used broadly. As an important aspect of sintering, it has been known that a small amount of additives which form a liquid phase during sintering can substantially enhance densification, i.e., liquid phase sintering (LPS), and result in a multiphase material. While a larger variety of materials are densified by LPS than solid state sintering (SSS), the fundamental study of the former is rather unsatisfactory mainly due to the complexity of the system, both in the number of phases present and in their geometry. Because of the geometrical complexity of these systems, Kingery's LPS theory has been criticized as it was based on a simple two sphere geometry.

Some materials cannot be densified successfully even with a small amount of liquid. Therefore, pressure sintering techniques such as hot pressing (HP) and hot isostatic pressing (HIP) have been used to accelerate densification and to control the microstructure more effectively than by conventional sintering. While it has been demonstrated in many materials that a dense, microstructurally controlled materials can be manufactured by pressure sintering, the role of the additional variable, pressure, is not satisfactorily understood. Thus, the full potential of this process has yet to be realized.

Therefore, to develop a more comprehensive understanding of multiphase material densification in the presence of a liquid phase, the objectives of this study were:

(1) To establish quantitative models to describe liquid phase sintering, and final stage densification by HIPing and liquid phase redistribution by HIPing of composite ceramics with a small amount of liquid phase.

(2) To analyze these models with respect to material properties and processing variables and to test the critical densification of multiphase ceramics.

(3) To determine the potential applicability of HIPing in densification of multiphase ceramics.

Summary

The present study mainly focused on the fundamental aspects of densification kinetics and microstructural development of materials in the presence of a small amount of liquid phase. Kinetic processes investigated include: (1) liquid phase sintering (LPS) under both pressureless and applied pressure conditions, (2) effect of gas diffusion and dissolution in a liquid during hot isostatic pressing (HIP), and (3) liquid phase redistribution by

capillary flow and macropore filling processes. Models have been developed for the respective kinetic processes. Effect of an applied isostatic pressure on the kinetics was investigated by HIP at less than 200 MPa. An alumina-glass system has been used throughout the experiments as a model system for ceramics which have a viscous boundary phase. Various experimental results have been analyzed based on the models presented. The results and conclusions are sumarized as follows:

1. LPS models were proposed. For modeling, a two-phase sintering map was developed on the basis of a quantitative geometrical analysis of the amount of liquid volume and sample density. A model for the rearrangement process was developed based on a viscous flow theory. A modified model for solution-precipitation was presented that incorporates a tetrakaidecahedron grain geometry that resulted in different particle size and time dependencies than previously proposed. Furthermore, these models account for variables, i.e., the effect of liquid volume and dihedral angle, that were not accounted for in previous models.

2. LPS densification data with an alumina-glass system was theoretically analyzed. Effect of the particle size, liquid volume, and activation energy measurements agree well with the proposed models for rearrangement and solution-precipitation. Activation energies measured were ~250 and ~500 KJ/mol for rearrangement and solution-precipitation, respectively. From various analyses, it is concluded that densification in the alumina-glass system is controlled by the interface reaction rather than diffusion. In the final stage of densification, samples exhibited a maximum in density and dedensification with extended sintering.

3. Final stage HIP densification models in the presence of a small amount of liquid phase were developed. An effective stress model was presented on the basis of a tetrakaidecahedron grain geometry which accounts for the liquid content and the dihedral angle. The model predicts that the solubility of the solid in the liquid at particle contacts significantly increases with applied pressure. The densification model presented is consistent with the high temprature creep-derived models previously reported.

4. HIP densification of alumina-glass composites sintered to ~95% density resulted in densities up to 99.5%. The effects of temperature, pressure, time, liquid content, and particle size on densification qualitatively agree with the models presented. However, extended HIPing at high temperature and pressure resulted in an end point density followed by dedensification. There were some difficulties in kinetic data analysis because of apparent data scatter as a result of the limited pressurization rate used and the possible gas diffusion effect during HIPing. Samples depressurized at a very high rate, $\geq$ 50 MPa/min, resulted in dedensification. Particle size dependence on densification, i.e., rs-1, is in agreement with pressure enhanced interface reaction control of solution precipitation.

5. A theoretical prediction of high pressure gas effects was made based on the collected gas diffusion and dissolution data in glasses and a diffusion model. The theory allows a satisfactory explanation for the gas diffusion effect on HIP densification. A series of annealing experiments with HIPed samples and glass resulted in sample swelling and corresponding dedensification, which supports the proposed gas diffusion effect. Design criteria for encapsulation materials in HIPing were proposed based upon these findings.

6. Theoretical models were developed for capillary flow of liquid through a porous powder compact by HIPing and for the pressure-assisted macropore filling. Applied pressure resulted in substantial pore removal by pressure enhanced capillary flow in the closed porosity alumina-glass composites as predicted by the model. Induced macropores were filled by pressure driven liquid and grain/liquid mixture flow into pores. The macropore filling behavior qualitatively agrees with the proposed model which accounts for the adhesive force between particles and capillary force in pores.

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