Characterization and Modeling to Control Sintered Ceramic Microstructures and Properties

Edited by C. DiAntonio

Characterization and Modeling to Control Sintered Ceramic Microstructures and Properties
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C.B. DiAntonio

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Preface

Predicting and controlling sintered ceramic microstructure and properties is a topic of considerable worldwide interest, and one that is becoming increasingly more important with multi-materials integration (e.g., microelectronics and other composite structures). Characterization, modeling and process control play an important role in the development and manufacture of traditional and advanced ceramics, particularly for process understanding and quality control/assurance. Practical characterization techniques are continually being developed and refined to better understand and control ceramic powder processing, and to support predictive modeling. Additionally, international collaborations are currently in place to develop practical characterization technology and international standards that will impact global ceramic manufacturing well into the 21st century.

This Ceramic Transactions volume is a collection of selected papers that integrates a variety of crucial areas in the development and understanding of sintering and densification. The papers include examinations into the characterization and modeling of sintered ceramic microstructures and the corresponding properties of those microstructures. This provides an increased understanding and expanded knowledge base that can be used by researchers and engineers. The papers were originally presented at the Characterization and Modeling to Control Sintered Ceramic Microstructure and Properties Symposium held during the 106th Annual Meeting of The American Ceramic Society in Indianapolis, Indiana, April 18-21, 2004. All of the papers in this volume were peer-reviewed.

The symposium was designed to provide a forum that integrated research in characterization and modeling to advance the science of ceramic/composite sintering. Densification, shape deformation, and microstructure evolution during sintering was addressed. Contributions on new developments and emerging characterization technologies, practical applications of traditional characterization techniques, and characterization techniques adapted from allied fields are presented. Three major topical areas emphasized in the symposium are presented in this collection: 1) characterization to support modeling (e.g., measuring constitutive properties) and to test and validate model predictions; 2) modeling to predict and control densification, shape deformation, microstructure, and properties; and 3) applications of characterization and modeling to engineer ceramic processes and ceramic microstructures.

I would like to personally thank Kevin G. Ewsuk, Sandia National Laboratories, and Eugene Olevsky, San Diego State University, for their assistance and encouragement in organizing this symposium. I hope that the dissemination of the work presented at the symposium, through this transactions volume, fosters a continued growth in the
understanding of the fundamentals as applied to sintering, characterization and modeling of ceramics. I would also like to acknowledge the numerous contributions and support of the speakers, conference session chairs, manuscript reviewers, manuscript authors and The American Ceramic Society officials. Your hard work and devotion to the advancement of science has allowed us to provide this volume to the ceramics community.

C.B. DiAntonio
Characterizing Sintering
Utilizing The Master Sintering Curve to Probe Sintering Mechanisms

D. Lynn Johnson
Department of Materials Science and Engineering
Northwestern University
Phone: 847-491-3584
Fax: 847-491-7820
Email: dl-johnson@northwestern.edu

Abstract

The master sintering curve (MSC) is empirically derived from densification data obtained over a range of heating rates or sintering temperatures. When the proper activation energy is used, all the data converge onto a single curve, the MSC. The activation energy can be estimated readily with just a few dilatometer experiments if it is unknown beforehand. Once established, the MSC makes it possible to predict the final density after arbitrary temperature-time excursions. It is particularly useful when considering alternative sintering methods. The MSC works if a single activation energy controls sintering, but also makes it possible to detect concurrent mechanisms.

Introduction

The ability to predict sintering behavior has been one of the long-term objectives of sintering studies for many decades. The complexity of the sintering process, which depends upon a large array of factors, largely has thwarted attainment of this objective. It is well understood that the sintering behavior of any given material depends upon several characteristics of the powder, including composition, the particle size and size distribution, particle shape, degree of agglomeration, agglomerate size and size distribution, and other factors. One of the principal composition issues is whether there will be a liquid phase present in the sintering temperature range. Compaction method, green density, and uniformity of green density are among the green compact issues that will influence sintering. Sintering temperature obviously has the most effect of any factor, but the heating rate may be an issue also. The atmosphere can have marked effects in some systems. A further complication arises if pressure is applied.

A predictive model would require knowledge of all relevant diffusion coefficients and their temperature dependencies, grain growth kinetics as a function of density, and more. If a liquid phase is present then the solubility of the majority material in the liquid, as well as liquid phase transport information must be known as functions of temperature. Thus, a truly predictive model, wherein sintering can be predicted based upon knowledge of all the relevant factors that might have a significant effect, has yet to be developed. Moreover, for practical purposes, such a model would require too much prior effort to acquire the necessary data. Thus, a more simplified approach is needed, even at the expense of detailed knowledge of every aspect of the process.

From the earliest quantitative sintering studies, over five decades ago, models have been sought that related the sintering rate to the characteristics of the particles, the compact, the atmosphere, and the temperature.[1-5] Simplified geometries were employed to readily identify driving forces, mass transport paths, and geometric factors as sintering proceeded. Attempts were made to extend these simple models into compacts of both spherical and non-spherical powders with limited success.

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Many of the simplified models can be derived from the following general model for densification by a combination of grain boundary and volume diffusion[6] by employing the geometric and other assumptions of the simplified models; the master sintering curve also derives from this model:

$$\frac{d\rho}{\rho \, dt} = \frac{3\gamma \Omega_a}{k_b \gamma} \left[ \frac{b D_b \Gamma_b}{G^4} + \frac{D_v \Gamma_v}{G^3} \right]$$  \hspace{1cm} (1)$$

where 
- $\rho$ = density 
- $\gamma$ = specific surface free energy, assumed to be isotropic (which, of course, it is not in general true for solids) 
- $\Omega_a$ = atomic volume 
- $k_b$ = Boltzmann constant 
- $G$ = mean grain diameter 
- $\delta$ = thickness of the region of enhanced diffusion at the grain boundary 
- $D_b$ = grain boundary diffusion coefficient 
- $D_v$ = volume diffusion coefficient 

$\Gamma_b$ = geometric factor for grain boundary diffusion = $\frac{a C_k C_b}{C_k C_a C_b}$  \hspace{1cm} (2)$$

$\Gamma_v$ = geometric factor for volume diffusion = $\frac{a C_k C_v}{C_k C_a C_h}$  \hspace{1cm} (3)$$

The geometric factors change continuously as sintering proceeds. They can be understood on the basis of the DeHoff model of a grain in a sintering compact.[7] Each grain is considered to be an irregular polyhedron defined by the grain boundaries between the grain and its nearest neighbors. The polyhedron, in turn, comprises pyramids with a common apex at the center of the grain, the bases of which are defined at the grain boundaries. The polyhedron is extended into the pores so that the total volume of the compact is included in the sum of all the polyhedra.

Each of the factors in Eqs. (2) and (3) relates a particular geometric factor for sintering to the mean grain size, $G$, as follows:

$$\nabla \mu = \frac{a K}{\lambda}$$

= gradient in chemical potential

$$\lambda = C_k G$$

= maximum distance of diffusion

$$K = - \frac{C_k^2}{G}$$

= curvature at the pore or neck surface

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