CONF-950634--7

SAND95-1371C

Experimentally Validated Computational Modeling of Organic Binder Burnout from Green Ceramic Compacts

K. G. Ewsuk, R. J. Cochran, B. F. Blackwell, J. Cesarano, and D. R. Adkins
Sandia National Laboratories, Albuquerque, NM 87185-1349

The properties and performance of a ceramic component is determined by a combination of the materials from which it was fabricated and how it was processed. Most ceramic components are manufactured by dry pressing a powder/binder system in which the organic binder provides formability and green compact strength. A key step in this manufacturing process is the removal of the binder from the powder compact after pressing. The organic binder is typically removed by a thermal decomposition process in which heating rate, temperature, and time are the key process parameters. Empirical approaches are generally used to design the burnout time-temperature cycle, often resulting in excessive processing times and energy usage, and higher overall manufacturing costs. Ideally, binder burnout should be completed as quickly as possible without damaging the compact, while using a minimum of energy. Process and computational modeling offer one means to achieve this end. The objective of this study is to develop an experimentally validated computer model that can be used to better understand, control, and optimize binder burnout from green ceramic compacts.

A combination of quantitative experimental analysis and computational modeling is being used to understand the complex phenomena associated with the burnout of organic binder from a powder compact. Experimentally, this entails utilizing conventional characterization methods (e.g., thermogravimetric analysis, TGA) as well as developing new techniques to quantitatively describe binder burnout. The experimental effort plays the critical role of providing the scientific foundation necessary to support model development. The goal of the computational modeling effort is to provide engineering level accuracy on inexpensive computer platforms with minimum analysis turnaround times for process design simulation.

To support the initial computational modeling effort, a one-dimensional (1-D) heat and mass transfer experiment was designed. Experimentally, 1-D radial heat flow was achieved using a rod-shaped heater that directly heats the inside surface of a stack of six ceramic annuli surrounded by thermal insulation. Type E thermocouples positioned in the ceramic stack were used to monitor radial and axial temperature gradients during heating. Mass loss and CO_2 emissions due to binder decomposition were monitored continuously during burnout using a computer interfaced balance and a mass spectrometer, respectively. A data acquisition system was used to record the time-dependent temperature, mass, CO_2 emissions, and power input to the heater.

Binder burnout experiments were conducted on a spray-dried 94 wt% Al_2O_3 powder containing 3 wt% of a 50/50 mixture (by weight) of methylcellulose and hydroxypropylcellulose binder. Ceramic annuli of approximately 6.7 cm outside diameter, 2.0 cm inside diameter, and 1.3 cm tall were formed by uniaxial pressing at 35-79 MPa, followed by isostatic pressing at 35-173 MPa. Burnout experiments were conducted on 57 and 65% theoretical density ceramic annuli stacked four high in the prototype, 1-D radial heat flow furnace. To minimize axial temperature gradients, annuli of low thermal conductivity zirconia insulation were placed on the top and

bottom of the Al_2O_3 ceramic stack, and the Al_2O_3 annuli in contact with the zirconia were pre-calcined (i.e. the binder was burned out before the experiment). Heating rates in the burnout experiments were varied by controlling the power input to the resistance heater. Power increases of 0.2 and 2.0 W/min. were used. Results from the burnout experiments confirm that 1-D radial heat flow is achieved in the prototype binder burnout furnace, and that detailed, highly reproducible data is obtained.

The computational modeling effort has centered on producing a macroscopic model of the burnout process. The models are based on continuum approaches to heat and mass conservation for porous media. The green compact is initially a porous body that is impermeable to gas flow. After calcining (i.e., binder burnout) the porous powder compact is permeable. The thermal decomposition of the binder is modeled using a set of increasingly complex computational models. The binder burnout process involves decomposition and combustion of the cellulose components that make up the binder. This process results in a heat and mass transfer problem involving both diffusion and convection processes in a porous matrix.

Currently, three levels of models are under development, including a no-flow, lumped-gas flow, and multi-component-gas flow model. The no-flow model is based on the energy conservation equation with heat conduction and an energy source term. The energy source term represents the energy release of the exothermic combustion reaction of the cellulose binder. Utilizing TGA data, a two-component Arrhenius reaction model was used to predict the mass loss of binder as a function of temperature. The mass loss was then used, with an assumed heat of reaction for the binder burnout reaction, to provide the magnitude of the energy source term as a function of time and temperature. This model predicts the temperature and mass of the green powder compacts as a function of time. The lumped-gas flow model introduces two mass conservation equations for ambient air and binder gas. The binder-gas is assumed to be the sum of the by-products of fully combusted binder. Diffusion of the two gas components are ignored in the mass conservation equations. Convection is added to the energy conservation equation to account for the gas flow in the porous matrix. The permeability is assumed to vary linearly as a function of the residual mass fraction of the binder present in the compact. This model adds the transient prediction of pressure inside the green compact sample, along with the concentration of the ambient air and binder gas in the porous matrix. The multi-component-gas flow model introduces mass conservation equations for the individual air components and the individual by-product components of binder combustion. A simplified multi-component diffusion model is used for the gas phase diffusion process in the mass transport model. This model introduces diffusion processes to examine the movement of oxygen, present in the ambient environment, into the reaction zone within the sample. Each of these models requires additional computer time to solve.

Results of the experimental studies are being used to test and validate the results of the computational models. Additionally, the computational models are being used to perform process rate studies. The simplest model that requires the fewest material property evaluations and minimum computational resources will be most desirable.

This work was supported by the United States Department of Energy under contract No. DE-AC04-94AL85000 at Sandia national Laboratories.

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.