Available online at www.sciencedirect.com

ScienceDirect

Ceramics International 41 (2015) 11870–11875

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Correlation between microstructure evolution and drying behavior of gelcast alumina green bodies

Xiang Peng[a,b], Shunzo Shimai[a,c], Yi Sun[a,b], Guohong Zhou[a], Shiwei Wang[a,*]

[a]The State Key Laboratory on High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, China

[b]University of Chinese Academy of Sciences, Beijing 100049, China

[c]Tokyo University of Agriculture and Technology, Tokyo 183-8538, Japan

Received 23 March 2015; received in revised form 11 May 2015; accepted 1 June 2015
Available online 6 June 2015

Abstract

Wet alumina green bodies with a dimension of 400 mm × 50 mm × 10 mm, respectively gelcast by PIBM (a copolymer of isobutylene and maleic anhydride) and EA (epoxy-amine) gel system, were dried at controlled temperature and humidity. Microstructure evolution and drying behavior of the green bodies with different solids loading and organic network were investigated. Pores among alumina particles became smaller and the constant rate period (CRP) became shorter with the increased solids loading or organic network. Further, the shrinkage of the body using PIBM ceased earlier and was smaller than that of the body using EA gel system. The typical microstructure of the body using PIBM gel system was thin organic networks on the particles and gradually a cocooned structure evolved during drying. While, the body using EA gel system had dense organic networks which evolved into a dense layer and strand-like structure around the particles. Such microstructures played different roles in water transportation and stress relaxation. As a result, the PIBM body was successfully dried without malformation but the EA body was bowing.

Keywords: A. Drying; Gelcasting; Microstructure evolution; Alumina

1. Introduction

Gelcasting [1,2] is the most promising way to manufacture large and/or complex ceramic components by means of in situ coagulation to create a homogeneous green body using simple equipment and processing techniques [3,4]. However, drying of a wet gelcast green body is particularly troublesome because water covers about half the initial volume of the green body, and it tends to warp and crack caused by the non-uniform capillary pressure in the small pores of the gel network [5].

Drying is such a field that has been frequently re-studied in the past several decades. Scherer et al. presented a classical review on the drying theory of pure gels (e.g. silica gel containing -Si–O–Si- 3D network and water) by a sol–gel processing [5]. For a gelcast ceramic green body, which is composed of ceramic particles and pure gel (organic network and water), the drying process was more complicated. On the one hand, the gelcast ceramic parts with a lower solids loading exhibited a higher drying rate using liquid desiccant drying method because solids loading significantly affected the drying potential [6,7]. On the other hand, Wang et al. gelcast alumina part by a modified free radical polymerization gel system, and demonstrated the graft chains upon heating could quickly release water through the skin layer formed during drying [8]. Moreover, Lewis et al. found that the gelcast alumina layer exhibited a complicated stress evolution because the PVA organic network was not beneficial for stress relaxation [9]. Furthermore, Ma et al. found that the organic network can be adjusted by a proper amount of hydroxyethyl acrylate so that the stresses were reduced in the ceramic green body [10]. For the first time, Ghosal et al. proposed a physical model which

*Corresponding author. Tel.: +86 21 52414320; fax: +86 21 52415263.
E-mail address: swwang51@mail.sic.ac.cn (S. Wang).

http://dx.doi.org/10.1016/j.ceramint.2015.06.001
0272-8842/

related particles with pure gels together, and assumed that the gel-matrix rupture happened due to the shrinkage of organic network around the particles. The rupture led to the change of channel for water transportation, and thus the drying process turned from constant rate period (CRP) to falling rate period (FRP) [11]. However, such a microstructure evolution has not been evidenced. It is inevitable that the microstructure, composed of solids loading and organic network, evolves during drying process for a gelcast green body. However, microstructure evolution and its influence on drying behavior have not been clarified so far.

In this study, we designed wet alumina green bodies with a dimension of 400 mm × 50 mm × 10 mm for one-dimensional drying and noticeable shrinkage detection during drying. They were gelcast using a copolymer of isobutylene and maleic anhydride (PIBM, hereinafter) [12,13]. For comparison, alumina green body was also cast by a epoxy-amine gel system [14]. The microstructure evolution of the wet green bodies during drying process was demonstrated, and its mechanism was proposed. The relationship between microstructure evolution and drying behaviors was discussed.

2. Experimental procedure

2.1. Materials and samples preparation

A commercial alumina powder (AES-11, Sumitomo, Osaka, Japan) with an average particle size of 0.45 μm was used as the raw material, which is shown in Fig. 1. PIBM with an average molecular weight of 55000–65000 (Isobam 104, Kuraray, Osaka, Japan), was used as both dispersant and gelling agent. The process for preparation of the alumina green body using PIBM was similar to a previous report [12]. Isobam 600AF from the same company with a molecular weight of 5500–6500 was added to increase solids loading [15]. Deionized water, alumina powder and 0.3 wt% PIBM (0.2 wt% Isobam 600AF + 0.1 wt% Isobam 104, relative to the weight of alumina powder) were mixed by ball-milling to make slurries containing 50–58 vol% solids. Slurries were degassed and cast into a mold with a typical size of 400 mm × 50 mm × 10 mm, and sealed by a plastic film to prevent water evaporation. The resultant alumina green body was noted as B-PIBM. The preparation of alumina green body using epoxy-amine gel system (noted as B-EA) was similar to that described by Mao et al. [14]. After gelling, the wet green bodies were dried in a commercial dryer (HWS-150, Sumsung, Shanghai, China) with a controlled temperature and relative humidity.

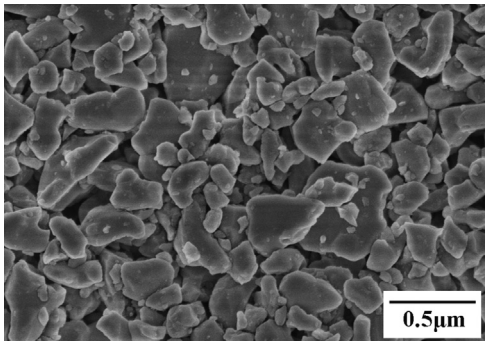

Fig. 1. Scanning Electron Microscope (SEM) image of the raw alumina powder.

2.2. Measurements

In situ drying loss (water removal) and shrinkage was measured by the modified commercial dryer mentioned above. An electronic balance was located on the top of the dryer to avoid the heat and humidity in the chamber (Fig. 2). A ruler was fixed to the sample supporter to measure the length of the sample. The supporter was tilted at 9 °in order to decrease the influence of the friction between the sample and the mold. The weight and length of the sample were recorded at 1 h intervals for the first day and at longer intervals on the following days. To measure the microstructure evolution during air drying, green bodies after different drying durations were firstly quenched in liquid nitrogen and then freeze dried. Microstructure of the green bodies on the fracture surface was evaluated using scanning electron microscopy (S-4800, Hitachi, Tokyo, Japan). Pore size distribution was measured using mercury porosimetry with a poremaster (PoreMaster-33, Quantachrome Corporation, Boynton Beach, FL).

3. Results and discussion

3.1. Influence of solids loading

The influence of solids loading on water removal of the B-PIBM was evaluated at a temperature of 40 °C and relative humidity (RH) of 60% (Fig. 3a). It's known that the rate of water vaporization (water loss) is the same in CRP stage because it only depends on the surface area and drying conditions. When solids loading increased from 50 to 58 vol %, the CRP duration of drying decreased from 20 to 10 h, and the volume fraction of residual water increased from 8% to 15% at the end of CRP. With further drying, the green body

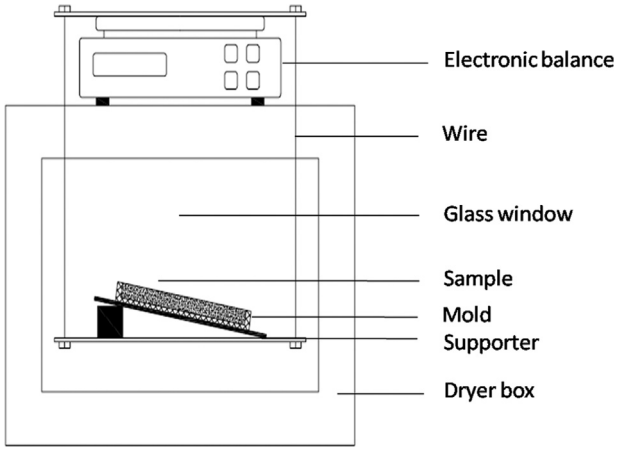

Fig. 2. Modified dryer for measuring drying loss and shrinkage.