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Cracking and shape deformation of cylindrical cavities during constrained sintering

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ABSTRACT

During constrained sintering of thin films, in which a cylindrical cavity with axis perpendicular to the substrate has been introduced before sintering, cracks emerge that initiate at the cavity surface. By combining experiments with continuum mechanical and particle based simulations, the fundamental causes and effects of this kind of crack formation are identified. A stress analysis performed by finite element (FEM) simulations matches with the cracking behavior observed in experiments. A comparison of discrete element (DEM) results with experiments shows the applicability of this simulation method to describe the effect of cross-sectional stripe dimensions and cavity diameters on the cracking behavior. Moreover, DEM simulations reveal that hair-line cracks in narrow stripe samples formed during pre-sintering manufacturing steps might be a dominant cause for the observed crack damage in such systems.

1. Introduction

Crack formation during sintering can be a significant problem in many applications of multi-material composites. For example, LTCC components as used in electrical, chemical or microfluidic systems are composed of multi-layered ceramics and metals to build up 3D circuits [1]. During manufacturing of the required thin films, agglomerates of organic material may form that are burned out prior to sintering. This can lead to the formation of internal voids or flaws at the surface of the film, which may be significantly larger than the particle dimensions [2]. On the other hand, internal voids can also emerge during co-firing if the layer material is a composite containing powder agglomerates with a higher sintering rate relative to that of the surrounding powder matrix [3,4]. Moreover, some voids are also introduced intentionally in the green films as defined cavities that later act as e.g. connection vias between the individual layers in a laminated multi-layer stack. In all of these cases it is important to know how size and shape of those voids develop during sintering and whether these internal surfaces are critical for crack formation and ultimately lead to failure of the part to function.

Only few experimental studies have systematically investigated the problem of crack initiation and development at such internal surfaces by introducing well-defined, artificial defects before constrained sintering. Bordia et al. [5] analyzed the damage and fracturing behavior in constrained sintered glass and alumina films of different thicknesses containing pre-existing, crack-like defects. They observed that crack growth occurs above a critical film thickness for the case of the glass film. The alumina film, however, just showed a diffuse damage zone ahead of the crack tip, so no conclusions could be drawn for crack growth in the alumina films. Cracks have also been observed by Green et al. [4], who presented an image of a circular via through a ceramic thin film after sintering, showing obvious damage in the form of cracks that are oriented radially around the void. However, this behavior was neither explained nor discussed in detail in this work. With the goal of optimizing the functionality of solid oxide fuel cells, Wang et al. [6] studied the defect evolution during constrained sintering of thin films composed of two different ceramic electrolyte materials. After binder burn-out they introduced artificial cracks in the films by Vickers indentation and observed a remarkable crack opening in the subsequent sintering step. Since no cracks at all appeared in the constrained sintered film when no artificial defects have been introduced before, they concluded that the existence of defects in the pre-sintering state is a necessary condition for cracking.

The problem of crack formation at pre-existing defects during constrained sintering was also investigated numerically in the past.

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Applying the so-called material point method, which uses a local empirical failure criterion and is based on continuum mechanical methods, Li et al. [7] simulated a simple system of a film with a rectangular defect. As a main result it was observed that all cracks develop at the corners, where the curvature of the defect is maximum. The first discrete element method (DEM) study about crack initiation during sintering was published by Henrich et al. [8], where a film composed of particles was constrained in one lateral direction and a notch has been introduced at the free film edge acting as a crack nucleation site. Because particle rearrangement was hindered by high inter-particle friction, a crack formed ahead of the notch tip and propagated through the whole sample in their simulations.

That inter-particle friction plays a crucial role for crack formation was also pointed out by Martin et al. [9]. Here, sintering of a thin film was simulated by DEM. The film was constrained by two opposing substrates in width direction, while free boundaries were used in thickness and the second width direction. Additionally, a crack-like defect was included in the system as an extra free surface, which passes through the whole film thickness and is situated in the interior of the film similar to the one in the experimental study of Bordia et al. [5] mentioned above. Applying a high friction coefficient, the pre-existing defect grows and new cracks form with direction perpendicular to the free edge. In contrast, sintering with frictionless particles did not lead to an effect on the initial crack at all. However, severe cracking behavior was also observed in samples that did not contain a pre-existing defect when inter-particle friction was high. On the other hand, sintering samples that contain a predefined defect but were not constrained in lateral direction lead to a closure of the defect. Therefore, these authors concluded that the geometrical constraint is a necessary condition for crack formation, but the existence of a predefined defect as a crack nucleation site has not been considered.

In contrast to the studies above, the intentionally introduced flaws in the thin film samples considered in this work are not crack-like or of an edge notch type. They are cylindrical cavities with major axis perpendicular to the rigid substrate that acts as sintering constraint. The samples are therefore similar to ceramic layers with inserted vias as they are used e.g. in ceramic multi-layer stacks for electrical and chemical applications. Due to the progress of miniaturization of mechanical, optical and electronic devices, a profound understanding about edge effects arising during manufacturing is compulsory. Therefore, not only films with approximately infinite width (continuous films), but also with finite lateral dimensions (stripe samples) have been investigated.

In this study, two different simulation approaches were employed besides an experimental investigation to analyze the cracking behavior in the vicinity of a cavity. The finite element method (FEM) based on continuum mechanics is the preferred method for calculating accurate stress distributions in short simulation times. In recent years, it has been successfully applied especially for the prediction of shape deformations during sintering [10,11]. However, the simulation of crack propagation is complicated with this method as it requires either complex remeshing and element deletion procedures or the use of advanced – and sometimes controversial – methods like XFEM (extended finite element method). Moreover, the continuum mechanical approach using FEM requires the application of a phenomenological damage initiation criterion. In our study, therefore, FEM has mainly been employed only for predicting the local stresses around cavities as well as geometrical changes of the cavity shapes, whereas the actual crack formation and propagation has been simulated by the particle-based DEM. Here, each grain of the ceramic powder is considered as a distinct particle that is able to move freely under conditions of interactions with neighboring grains. As a consequence, this method is much more computation time consuming, but the effect of crack initiation is included by design.

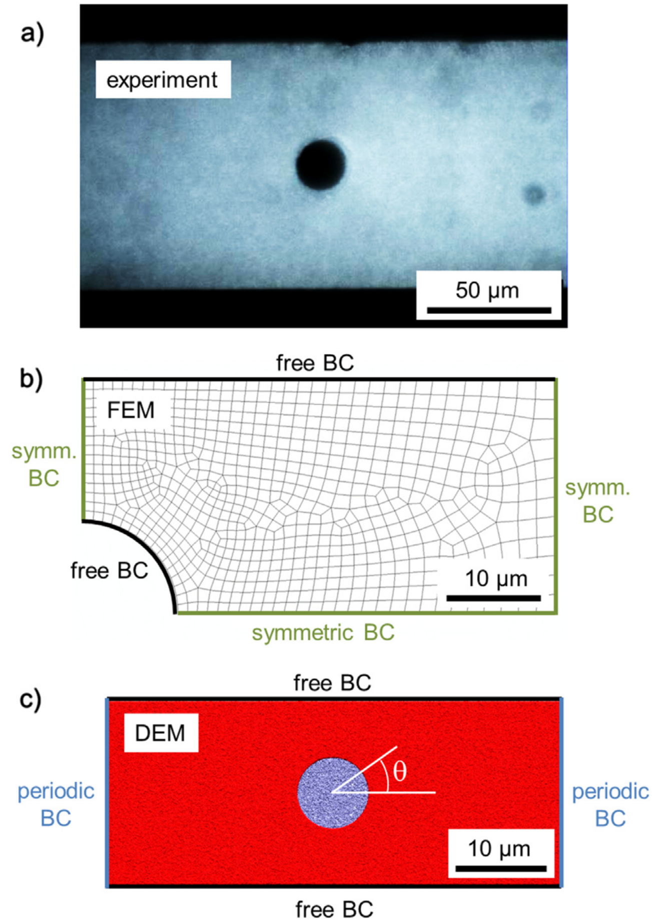

Fig. 1. Top view on examples of green stripes on a rigid substrate including a cylindrical cavity in experiment (a) and as used for FEM (b) and DEM calculations (c). The DEM sample consists of 2.35 Mio. particles (red particles represent the film, blue particles represent spatially fixed substrate particles).

In the following, the applied simulation methods and the experimental procedure for solid state sintering of thin films on a rigid substrate are introduced. Results for all of the three different methods are compared with each other, with a focus on the cracking behavior at the cavity for stripes with variable geometry. Finally, the findings are discussed with regard to the origin of the observed effects and to discrepancies between the outcomes of the different methods.

2. Experiment and simulation description

Preparing appropriate green stripe samples with predefined cavities for sintering experiments and simulations is a challenging task. While the manufacturing of micro-sized and standalone samples with high aspect ratios on a substrate requires sophisticated and elaborate experimental methods like soft lithography, the difficulty of initial sample design for simulations is mainly about finding an appropriate choice of boundary conditions and a suitable dimensioning of the sample in order to keep the computational costs within a reasonable limit. Examples of green samples for the individual methods employed in this study are shown in Fig. 1. Manufacturing details and modeling concepts will be described in detail in the following subsections.