



Effectiveness of wick-debinding inside powder bed for ceramic laminates made by tape casting



Lovro Gorjan, Gurdial Blugan, Thomas Graule, Jakob Kuebler

Empa, Materials Science and Technology, Laboratory for High Performance Ceramics, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland

ARTICLE INFO

Article history:

Received 12 October 2014
Received in revised form 20 December 2014
Accepted 28 December 2014
Available online 5 January 2015

Keywords:

Tape casting
Debinding
Capillary extraction
Master decomposition curve

ABSTRACT

Laminated Al₂O₃ samples were prepared by tape casting and warm pressing of multiple layers into compact parts. The effect of a highly porous powder bed on the debinding process has been investigated by performing different debinding cycles with or without the powder bed. The effectiveness of the powder bed was analyzed using the concept of master decomposition curve. Results show that the activation energy of the binder removal process is significantly lowered when the debinding takes place inside the powder bed due to capillary extraction effects. Critical defects such as bloating and cracking are also avoided. In order to further exploit the capillary extraction phenomenon for the debinding a modification to the binder system was made, where a low-melting point component was added, which resulted in improved effectiveness of the capillary extraction.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Removing the organic binder from green ceramic parts before the sintering is considered a time consuming and delicate operation for certain shaping techniques where the green part contains large amount of binder as in tape casting and lamination. Several green ceramic tapes can be stacked and warm pressed to shape a thick multi-layer green body [1–3]. Since ceramic tapes are made from suspensions that contain large amount of organic binders and plasticizers, the green body therefore contains a large amount of organics (up to 50 vol.%) that must be removed prior to sintering without causing any disturbance to the compacted body. Usually a thermal debinding process is applied, during which green bodies are slowly heated until the binder decomposes and leaves the body in the gaseous form [4]. This is a critical step, because serious flaws may be introduced. If the decomposition of the binder during heating is too rapid, then a high pressure can be built-up in the interior of the green body and cause damage such as cracks and delamination [4,5]. To avoid such defects, slow heating cycles are used that can last over one hundred hours and thus present the longest step in the production [4,6]. In order to reduce the debinding time, optimization of the thermal debinding program can be achieved by studying the kinetics of the binder decomposition [4–8] or to introduce additional assistive process like debinding in the centrifugal field [9], partial debinding with supercritical carbon dioxide [10–12] or debinding with microwave heating [13]. However one particularly effective and simple way of reducing the formation of defects and shorten the debinding time is to introduce an additional debinding step that takes place inside a capillary extraction embedment, which has been extensively studied for the powder injection molding [14–22]. The green body is embedded in a highly porous powder bed and is slowly heated.

When the binder or part of the binder melts, it is sucked into the fine pores of the embedment by capillary forces. This process offers a fundamentally different mechanism of binder removal, compared to the common thermal debinding. The basic difference is that the binder leaves the green body in the liquid phase and so it can be removed very rapidly without the risk of pressure build-up. The powder bed also offers a gentle physical support for the green parts, regardless of their shape, and thus helps to prevent certain flaws, such as shape distortion and cracking [14].

The progress of binder removal can be studied by using the idea of master debinding curve (MDC). The concept was first developed by Su and Johnson [23] as a master sintering curve (MSC) for describing the densification during the sintering and was later successfully applied for various ceramic [24–26] and metal [27–29] powder systems. The concept is very general and it turned out that it can be successfully used also for other thermal processes like thermal debinding, solvent debinding and also for the wick-debinding of PIM samples [30]. The central part of the MDC idea is the special Θ -function (Eq. (1)) which summarizes the effect of thermal history of the sample during the debinding process. The relationship between the binder content and the Θ -function is called master debinding curve and it is the unique curve for a certain green body composition, regardless of the time–temperature cycle. Therefore, it can be used as a useful tool for optimization, since only a limited number of experiments give enough data to predict an arbitrary debinding cycle.

$$\Theta(t, T(t)) = \int_0^t \frac{1}{T} \exp\left(\frac{-Q}{RT}\right) dt \quad (1)$$

where t [s] is time, T [K] is temperature at the time t , R [$8.31 \text{ J mol}^{-1} \text{ K}^{-1}$] is universal gas constant and Q [kJ mol^{-1}] is the activation energy of the process and is the only material parameter in the equation. If the MDC is applicable for a certain system (green body) then the activation energy value (Q) can be found for which all experimental data (for different time–temperature cycles) merge into a single curve on a graph where binder content vs. $\text{Log}(\Theta(t, T))$ is plotted. The determination of the activation energy (Q) is achieved numerically by altering the value of Q until all experimental data best fit the single curve.

The motivation for our work was to assess the effectiveness of the capillary extraction for the debinding of green parts made by the tape casting and lamination. In this paper we present experimental result showing the effect of capillary extraction powder bed for two different polyvinyl butyral based binder systems. Many different heating cycles were tested in order to construct MDC and evaluate the activation energy of the debinding process. Moreover, by modifying the binder formulation with capillary extraction in mind, increased effectiveness is expected.

2. Experimental materials and methods

Fine ($d_{10} = 0.2 \mu\text{m}$, $d_{50} = 0.4 \mu\text{m}$, $d_{90} = 1.2 \mu\text{m}$) high purity alumina (Ceralox HPA 0.5 doped with 500 ppm MgO; Sasol North America Inc., USA) was used as the starting powder for fabrication of samples by tape casting. Two different formulations were used – marked as “feedstock A” and “feedstock B”. For feedstock A a polyvinyl butyral (PVB) Mowital B30 H (Supplied by Omya AG, Switzerland) based binder system was used, with ethanol as a solvent, polyethylene glycol PEG 300 (Fluka Chemie GmbH, Germany) as a plasticizer and Solsperse 41000 (Lubrizol Deutschland GmbH, Germany) as a dispersant. Such a binder system has already been proven for tape casting and lamination [31]. For the feedstock B the modified binder system was used with the aim to further improve the effectiveness of the capillary extraction. The idea behind formulation of feedstock B was to decrease the amount of PVB which does not melt into a low viscosity liquid and replace it with the PEG 3000 (Fluka Chemie GmbH, Germany) which is solid at room temperature, but melts into low viscosity liquid. The detailed formulations for the feedstocks are presented in the Table 1, where m is absolute mass of component, w is weight percent, vol. is volume percent and vol. dry is volume percent of a certain component in the dried tape and also in the green sample after lamination.

The processing for both suspensions was the same. First, the surfactant was dissolved in the ethanol inside a 250 ml polyethylene bottle and then the powder and $\Phi 3$ mm yttria-stabilized zirconia milling balls (SiLibeads type ZY, Sigmund Lindner GmbH, Germany) were added. The suspension was homogenized on the roller mill for 6 h. After that PEG 300, PEG 3000 (in the case of feedstock B) and half of the amount of PVB were added (4.5 g in the case of feedstock A and

2.25 g in the case of feedstock B). After another 2 h of homogenization the rest of the PVB was added. The suspension was ready for tape casting after an additional 24 h of homogenization. The last 2 h of homogenization was done with slow rotational speed in order to remove air bubbles from the suspension.

Tape casting on a polyester film substrate with a siliconized surface (Rostaphan® RN 75 SLK from Mitsubishi Polyester Film GmbH) was done with a single doctor blade machine. The gap between the doctor blade and the substrate was set to 0.60 mm and the speed of the doctor blade was 0.5 m/s. Tapes were dried overnight and then removed from the substrate. Squares were cut from the dried tapes, 10 layers were stacked on top of each other and warm-pressed in a 20×20 mm steel die for 5 min with 25 MPa of pressure at 70°C . The thickness of the green samples was 1.60 ± 0.05 mm.

The debinding experiments were performed in a ventilated oven (Heraeus UT 6060) up to 300°C . For each parameter two different kinds of samples were tested; one was placed directly in metal container and the other was embedded in a calcined alumina Nabalox NO 201 powder bed (from Nabaltec AG, Germany). The powder bed was in the form of coarse porous particles with d_{50} of $80 \mu\text{m}$ and BET specific surface area of $75 \text{ m}^2/\text{g}$. Two different types of debinding cycles were used for experiments:

- Linear heating cycle, characterized by the constant heating rate. Heating rates of 0.1, 0.2, 0.5, 2.0 and 3.0 K/min were used in the experiments.
- Isothermal heating cycle, characterized by the constant temperature. Dwell temperatures of 100°C , 120°C , 130°C and 140°C were used in the experiments.

After a certain amount of time in the oven the samples were removed, cleaned and weighed. The weighing was done with an analytical balance (Mettler Toledo XSE205). Thermal analysis was done with Mettler Toledo TGA/sDTA 851e using the heating rate of 5 K/min in air atmosphere. Pore size distributions were measured by Pascal 140/440 mercury porosimeter.

3. Results and discussion

Results of thermal analyses of both feedstocks are shown in Fig. 1. A distinct endothermic peak can be seen in the case of feedstock B, which is a consequence of the melting of the PEG 3000. The degradation of both feedstocks is quite similar; an intensive exothermic decomposition starts at around 170°C and progresses until 500°C when all of the binder decomposes. Feedstock A undergoes slower and more gradual degradation.

Table 1
The composition of feedstocks used for the tape casting.

		m [g]	w [%]	Vol. [%]	Vol. dry [%]
Feedstock A					
Powder	Al_2O_3 (Ceralox HPA 0.5)	100.0	60.2	24.6	58.1
Dispersant	Solsperse 41000	2.0	1.2	1.8	4.2
Binder	Polyvinyl butyral (Mowital B30 H)	9.0	5.4	8.0	18.9
Plasticizer	PEG 300	9.0	5.4	8.0	18.9
Solvent	Absolute ethanol	46.0	27.7	57.6	/
Feedstock B					
Powder	Al_2O_3 (Ceralox HPA 0.5)	100.0	60.6	24.9	58.1
Dispersant	Solsperse 41000	2.0	1.2	1.8	4.2
Binders	Polyvinyl butyral (Mowital B30 H)	4.5	2.7	4.1	9.4
	PEG 3000	4.5	2.7	4.1	9.4
Plasticizer	PEG 300	9.0	5.5	8.1	18.9
Solvent	Absolute ethanol	45.0	27.3	57.1	/

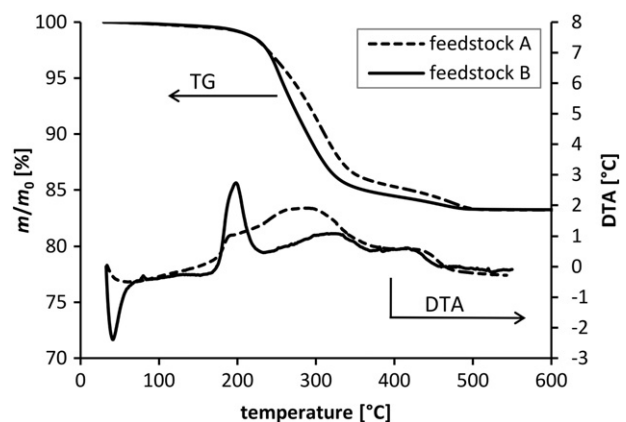


Fig. 1. Results of thermogravimetric analysis (TG) and differential thermal analysis (DTA) of feedstocks.

Download English Version:

<https://daneshyari.com/en/article/235648>

Download Persian Version:

<https://daneshyari.com/article/235648>

[Daneshyari.com](https://daneshyari.com)