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Drying of a tape-cast layer: Numerical modelling of the evaporation process in a graded/layered material

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ABSTRACT

Evaporation of water from a ceramic layer is a key phenomenon in the drying process for the manufacturing of water-based tape cast ceramics. In this paper we present a coupled free-flow-porous-media model on the Representative Elementary Volume (REV) scale for coupling non-isothermal multi-phase compositional porous-media flow — for the ceramic layer — and single-phase compositional laminar free flow — for the air above it. The preliminary results show the typical expected evaporation behaviour from a porous medium initially saturated with water, and water–vapour transport to the free-flow region in accordance with the available results from the literature. We elaborate on and discuss the characteristic drying-rate curve for a single layer ceramic, and compare it with that of a graded/layered ceramic. We, moreover, show the influence of the mean diameter of particles of the porous medium (d_p) — which directly affects the intrinsic permeability ($\mathbf{K}$) based on the well-known Ergun's equation — of each single ceramic layer on the drying behaviour of a graded/layered ceramic.

1. Introduction

Tape casting is a well-established technique (cf. Fig. 1) in producing thin ceramic layers, electronic substrates and multilayer ceramics (MLCs) [1–3]. Due to the nontoxic nature of the fluid vehicle, availability, environmental friendliness, noncombustibility, and low-cost of production [4], the use of aqueous-based slurries is growing in the tape casting process. However, these types of slurry systems have not been matriculated by industry because of problems related the drying process, e.g. longer times for drying and higher probability of defects [5].

The drying stage and the characterization of it in the form of final shrinkage is often measured experimentally, simply by the weight difference of the green and dried tapes as well as "in situ" weight-loss measurement, without really noticing that the drying is one of the most important steps in the tape casting process. Drying often contains different kinetics, i.e. evaporation, viscous deformation, flow in porous media, and diffusive transport [6–8], whose coupled behaviour influences the final properties of the thin layers.

The ceramic slurry used in the tape casting process contains different ingredients, i.e. solvent, dispersant, binder, plasticizer and deflocculant, each of them having a specific influence on the rheological behaviour of the slurry and consequently the final properties of the tapes. This has been studied both experimentally [9,10] and numerically [11–14] in literature. Hence, a tape layer can be considered as a porous medium which contains powders and liquid phases [4].

Multilayered ceramic circuits/packages (MLCCs) would not exist if tape casting had not been invented [15]. The basis for the multilayer products is the ability to individualize layers with respect to metallization and via interconnections and then to laminate a set of these individual layers together into a package that can be sintered into a monolithic structure. Multilayered ceramic packages with as few as two layers up to structures with as many as a hundred or more layers are common in the electronic ceramics industry today [16]. Moreover, multilayered ceramics produced by tape casting, have also been developed and used for different applications, i.e. flue gas purification [17] and magnetic refrigeration [18,19], thus underlying the growing diversity of today's applications of the tape casting process. Apart from the MLCs, typical applications of multilayered (non-uniform) material include the tertiary oil recovery process, geothermal analysis, asphalt concrete pavements process and preservation process of food items. Therefore, knowledge of the heat and mass transfer that occurs during

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Nomenclature

$\widehat{\mathbf{K}}$	average permeability, m^2
p_c	capillary pressure, Pa
M^κ	component molar mass, K/mol
D	diffusion coefficient, m^2/s
$\dot{R}$	drying rate, mMol/min
$\mathbf{g}$	gravity vector, m^2/s
c_s	heat capacity of porous matrix, J/kg·K
H_{gl}^a	Henry's law constant, Pa
$\mathbf{I}$	identity matrix
$\mathbf{K}$	intrinsic permeability tensor, m^2
L	length, m
$\mathbf{F}$	mass flux, kg/m^2 s
$y_{\max}^{ff}$	maximum y in free-flow domain, m
$v_{\max}$	maximum velocity in free-flow domain, m/s
d_p	mean particle diameter, m
$y_{\min}^{ff}$	minimum y in free-flow domain, m
$\mathbf{n}$	normal vector
u_α	phase internal energy, J/kg
X_α	phase mass fraction
x_α	phase mole fraction
p	phase pressure, Pa
h_α	phase specific enthalpy, J/kg
$k_{r\alpha}$	relative permeability
Re	Reynolds number
S	saturation
q	source or sink term
T	temperature, K
t	time, s
$\mathbf{v}$	vector of phase velocity, m/s
v_x	velocity in free-flow domain, m/s
Q	volumetric flow, m^3/s
W	width, m

Greek symbols

α_{BJ}	Beavers–Joseph coefficient
ϱ	density, kg/m^3
Ω	domain
λ_{pm}	effective lumped heat conductivity, W/m·K
Γ	interface
$\lambda_{eff,\alpha}$	phase effective heat conductivity, W/m·K
μ_α	phase viscosity, kg/m·s
ϕ	porosity
τ	tortuosity

Subscripts

g	gas phase
l	liquid phase
α	phase
pm	porous medium

Superscripts

a	air component
κ	component
ff	free flow
pm	porous medium
w	water component

Acronyms

MLC	multilayer ceramic
MLCC	multilayered ceramic circuit
REV	Representative Elementary Volume

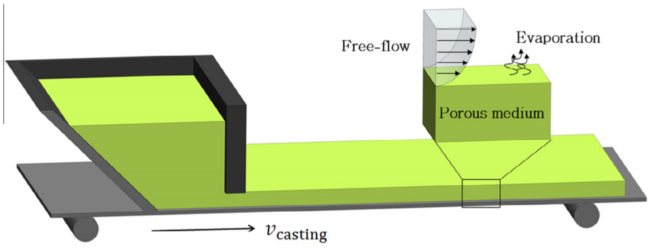

Fig. 1. Schematic illustration of the tape casting process.

drying of graded/layered porous materials is necessary to provide a basis for fundamental understanding of convective drying of non-uniform materials [20,21].

The prediction of the evaporative drying rate from a porous medium remains a challenge due to the ambient conditions at the interface (radiation, humidity, temperature, air velocity, turbulent conditions) [22]. Moreover, the internal porous-media properties lead to abrupt transitions in flux dynamics [23,24]. The factors involved are coupled by the complex interactions between the porous medium and the free-flow system [25,26].

Modelling such coupled systems while accounting for the respective processes in both domains is a challenging task, especially since many of these systems are dominated by multi-phase compositional flow and moreover using different model concepts [22]. A non-isothermal two-phase compositional flow model in the porous medium being in contact with a laminar single-phase non-isothermal compositional system in the free-flow region, was initially developed by Mosthaf et al. [27] to capture the evaporation phenomenon in environmental systems. The employed coupling conditions for mass, momentum and energy are valid on the REV scale, and account for the physics at the interface. These conditions are based on flux continuity and local thermodynamic equilibrium at the interface and are justified by phenomenological explanations.

The focus of this paper is to simulate the evaporation phenomenon in a graded/layered ceramic. For this purpose we present a comprehensive model similar to the work developed by Mosthaf et al.[27] and further studied in [22,28–30]. However, the current study focuses on developing a model for a smaller scale (size), a different relationship for the intrinsic permeability of the porous medium as well as a different configuration for graded and layered structures. Due to the limited amount of experimental work on drying of multilayered materials, the various effects are not fully understood and a number of critical issues remain unresolved. The effects of particle size and the layered configuration on the overall drying kinetics have not been systematically studied. Although most previous investigations consider single-layered material, little effort has been reported on convective drying of multilayered material (non-uniform structure) at a fundamental level [20,21]. This will be studied thoroughly in this paper. In the following, the developed model concept will be briefly explained together with the implementation into the modelling toolbox DuMu^X [31].

2. Mathematical model

The model concept used in this study combines a two-phase flow (gas and liquid) in the porous medium, and a single-phase