



## A model for the non-isothermal drying of particle-laden pastes



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### ARTICLE INFO

#### Article history:

Received 19 December 2014

Received in revised form 2 September 2015

Accepted 3 September 2015

Available online 21 September 2015

#### Keywords:

Drying

Paste

Particles

Sedimentation

Hindrance function

PVA–water

### ABSTRACT

A one-dimensional theoretical model for the non-isothermal drying of particle-laden pastes on solid substrates is developed and solutions of the model transport equations for the mass fractions of the liquid mixture components, the particle volume fraction and the thermal energy are presented. The aim of the drying process is to produce a homogeneous layer of particles embedded in a binder matrix with a controlled microstructure. The shrinkage of the liquid layer due to the solvent evaporation may be either faster or slower than the particle sedimentation. Numerical solutions of the transport equations subject to initial and boundary conditions for an industrial process show that the former case prevails here. The results are particle packings with variable particle volume fractions, which may change with both the initial and the drying conditions. A guideline for proper selection of the initial and drying conditions ensuring a homogeneous particle packing emerges.

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### 1. Introduction

In many industrial applications, pastes laden with particles are used for producing layers consisting of immobilised particles on substrates. After application of the paste on the substrate, the paste is cured – which often consists of a simple drying step as considered further in this study – leaving the particles embedded in a binder matrix. This process, commonly called thick film technology, can be used for producing functionalized films with a wide range of properties conferred by the nature of the embedded particles.

To name a few, conductive, resistive and dielectric materials including ceramic have been widely used in the printed circuit board industry [1], in the sensing industry especially for producing electrodes [2–4] or – more recently – in the packaging industry producing resonant tags for electronic article surveillance [5]. Layers obtained from magnetic particles can be used as inductors [6] for printed electronics or as magneto-resistive materials for magnetic sensing [3]. Semiconductors for transistors [7] and photovoltaics [8], as well as complete photovoltaics cells [9], can be produced using thick film technology. The technology has recently been extended to electrochromic materials and organic

light emitting materials which appeared to be very promising for producing various kinds of display, including flexible ones [10]. For a few decades already, a considerable number of commercially available electrochemical sensors have been produced applying successive layers ensuring various roles from electrode to porous reservoir or selective membranes as shown in the reviews [11,12].

Independently of the method used for paste deposition, for any of these applications the performances of the layer are directly related to its thickness, its homogeneity and the packing of the embedded particles. More precisely, the particle volume fraction and its spatial distribution within the package are crucial [13]. Yet, to our knowledge only limited research has been published about the effects of drying conditions of a paste on the microstructure of the resulting materials. Only a few studies treated the case of non-Brownian particles [14,15], while most of the work deals with colloidal suspensions [16–19] excluding *de facto* particle sedimentation, whereas it is essential in most commercially available pastes, since they contain non-Brownian particles.

The behaviour of the drying pastes may be characterised according to the dominant particle behaviour. In the general case, particles may move due to settling induced by gravity and due to Brownian diffusion. The relevant diffusion coefficient is given by the Stokes–Einstein equation

$$D = \frac{kT}{3\pi\mu d_p}, \quad (1)$$

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where  $k$  is the Boltzmann constant,  $T$  the system temperature,  $\mu$  the liquid dynamic viscosity and  $d_p$  the particle diameter. In the context of stabilized particles dispersed in a paste at rest, the Brownian character of the particles can be defined using a Peclet number  $Pe_{sed}$  based on sedimentation and thermal agitation time scales for the convective and diffusive terms, respectively [14,15,20]. We define this Peclet number different from [14] in using the particle diameter  $d_p$  as the typical length scale

$$Pe_{sed} = U_0 d_p / D = \pi(\rho_p - \rho_m) g d_p^4 / 6kT, \quad (2)$$

where  $U_0$  is the Stokesian settling velocity of an individual particle in the paste,  $\rho_p$  and  $\rho_m$  are the particle and liquid densities, respectively and  $g$  is the gravitational acceleration. Using typical values of commercially available pastes, such as  $d_p \approx 2 \mu\text{m}$ ,  $\rho_p - \rho_m \approx 10,000 \text{ kg/m}^3$ , and considering wet-bulb temperatures in the range of 400 K leads to  $Pe_{sed} \sim 160$ . In this limit of large  $Pe_{sed}$ , particles are by definition non-Brownian and will be considered as such in the rest of this study.

The particle settling and diffusive motions may also be compared to the shrinking motion of the free surface due to evaporation, which results in two different characteristic numbers. The two emerging ratios of velocities may be expressed by the particle Peclet and sedimentation numbers [14]

$$Pe = \frac{\dot{h}}{D/d_p} \quad \text{and} \quad N_s = \frac{U_0}{\dot{h}}, \quad (3)$$

where  $\dot{h}$  is the rate of shrinkage of the layer thickness and we again used the particle size as the relevant length scale. According to the numerical values of these numbers, drying regimes may be identified where the particle settling, the solvent evaporation or the particle diffusion dominate the behaviour during drying [14]. Oriented at these criteria, drying regime maps were presented in [14]. As derived above, the systems in our study are non-Brownian and exhibit large values of the  $Pe$  number. Typical values of  $Pe$  in our study range well above  $10^3$ .

The aim of the present work is to gain understanding of the drying process of non-colloidal particle-laden pastes in connection with the resulting film thickness, film homogeneity and particle packing. We therefore restrict our study to applications for which curing conditions consist of a simple drying step. The focus is on the parameters with the highest influence on the particle distribution. The pastes used in industry are mostly proprietary, thus their properties and underlying recipes are not public. In the present work, a model liquid phase is treated as a binary mixture of a solvent and a solute. The test system of poly(vinyl alcohol) in water was used in the calculations. The goal of the simulations is to compute time-dependent concentration profiles and temperatures for both the solid and liquid phases as well as profiles of the particle volume fraction inside the layer at specific drying conditions. These results enable the analysis of the produced layers. The thickness of the layer and the uniformity of the particle volume fraction distribution are investigated at the end of the drying process. The most influential material and process parameters are identified, and guidelines for achieving the densest and most uniform packings are provided.

The paper is structured as follows: Section 2 formulates the problem and describes the governing transport equations for both the liquid and the solid phases. Furthermore, it details the applied physical models and the calculation procedure and provides the material and process parameters for the calculations. Section 3 presents the validation of the modelling and computational approach and shows the results from the drying simulations. The paper ends with the conclusions.

## 2. Mathematical description

### 2.1. Specification of the problem

The drying of a thin particle-laden liquid layer on a planar substrate is investigated. The liquid is a binary mixture of the solute poly(vinyl alcohol) with the solvent water. The initial solvent mass fractions vary between 0.6 and 0.8. It contains a disperse phase of solid metal particles with initial volume fractions between 0.23 and 0.31. The viscous behaviour of the liquid justifies terming it a paste. The drying is driven by an air flow above the layer with a well-defined thermodynamic state.

For quantifying the drying process, the mass transport in the liquid phase and across the free layer surface is analysed. In the liquid phase, the model accounts for the transport of the solvent by diffusion and for a convective motion induced by non-equimolar diffusion, as well as by the particle settling. Due to the application we have in mind, we neglect diffusion in the modelling of the particle motion as laid out above. The particle settling is driven by gravity and hindered by the hydrodynamic drag. The latter is due to the liquid viscosity and influenced by the presence of other particles in the suspension, which results in a mutual hindrance of the particles in the settling process. Hence, to analyse the drying process, a coupled system of transport equations for the liquid phase with the particles must be solved. The Cartesian geometry of the system, with the  $x$  axis pointing upward normal to the plane of the liquid layer, and the transport phenomena accounted for, are depicted in Fig. 1.

Simple power laws for the non-dimensional heat and mass transfer coefficients on the gaseous side were selected as the models. In this approach, the Sherwood number  $Sh$  was formulated as a constant times the Reynolds and the Schmidt numbers to the power of some numbers, and the Nusselt number  $Nu$  for the related heat transfer process was derived from  $Sh$  by the Lewis analogy equation [21]. Both the constant and the exponent were determined in a set of experiments covering the range of drying air velocities and temperatures relevant to the real production process. The mass loss measured in the experiment is due to the solvent evaporation only. The dissolved polymer (the solute) has a negligible vapour pressure and remains in the layer. It binds the particles together and fixes their positions in the layer. Producing a layer of immobilised particles on a substrate is the aim of the process studied.

The glass substrates and the liquid layers are  $150 \mu\text{m}$  and  $60 \mu\text{m}$  or  $960 \mu\text{m}$  thick, respectively, and the values of the heat transfer coefficient and the thermal conductivities of the materials are such that the resulting small Biot numbers  $O(10^{-3})$  lead to flat temperature profiles in the substrate and the paste layer. Moreover, the high Lewis number  $Le \sim O(10^3)$  in the paste layer means that heat conduction is significantly faster than mass diffusion. Consequently, it can be assumed that the temperature of the liquid layer is uniform. Moreover, assuming negligible thermal contact resistance between the paste layer and the substrate, the liquid temperature may be assumed equal to the temperature of the substrate at all times. The evolution of the liquid phase temperature with time may therefore be modelled by a differential heat balance equation following the lumped capacitance approach, as described in [22]. The effect of the particles on the heat-up of the paste material is accounted for by including their heat capacity. Yet, the metal particles, having larger heat conductivity than polymers, raise the Lewis number of the paste. The particles therefore enhance the uniformity of the temperature of the paste, while the mass diffusivity of the solvent is not affected.

The mass transport in the liquid film has three main driving mechanisms. The first one is diffusion due to concentration

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