

Modelling the shrinkage in pigmented coatings during drying: A stick–slip mechanism

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Abstract

Pigmented coatings, used to improve optical and printing properties, are applied to fibrous paper substrates as slurry, which then dries. We have elucidated the mechanism of the shrinkage which occurs during drying. The void space of the dry coating layers and their effective solid skeletal elements were modelled using the porous network simulation software Pore-Cor. The water-filled porous structures at the beginning of the shrinking process were modelled by creating simulated structures with the same effective skeletal element size distribution as the dry ones, but with higher given porosity to account for the water present. The capillary forces acting on the surface of the drying coating were calculated for the model structures and found to be orders of magnitude larger than the experimentally measured shrinkage forces. The shrinkage process was therefore postulated as resulting from the effect of capillary forces resisted by a discrete stick–slip process. The differences in the visco-elastic properties of the slurries also supported this postulate, as did further experimental evidence.

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1. Introduction

The application of a pigmented coating layer to a base paper substrate improves its optical, coverage and printing properties such as uniformity in appearance, gloss, opacity, ink absorption, and spread.

A pigmented coating layer contains pigments such as ground or precipitated calcium carbonates, clays, polymeric pigments (such as polystyrene), titanium dioxide, silica, or talc. It also contains binders, which are needed to provide good cohesion of the porous structure formed by the pigment and adhesion of this structure to the substrate.

A commercial coating ‘colour,’ a suspension of pigment and binder in water, has a solid content of between 50 and 70 percent by weight (wt%). This solid content comprises 80–90 wt% of pigment, and 10–20 wt% of binder. Other compounds, such as dispersants, are used in lower percentages to act as stabil-

ising agents and to make the components compatible in water suspensions. The coating colour is applied onto the substrate and is usually dried thermally using infra-red and hot air dryers.

Watanabe and Lepoutre [1] divide the drying process into three phases, separated by two critical concentrations (Fig. 1). The slurry or coating colour dries until it reaches the first critical concentration (FCC), at which a three-dimensional fluid-filled network is formed and particle motion is greatly restricted. In the second phase, the water-air interfaces recede into the surface pores forming capillary elements, creating a differential capillary pressure that induces a shrinkage of the network. This continues until the second critical concentration (SCC) is reached, at which the structure is rigid and air enters as the liquid retreats. In this work we introduce an inter-critical concentration (ICC), which is an intermediate stage between FCC and SCC, as described in Section 4. It is the structure ranging from FCC to SCC that is important with regard to shrinkage forces—before the FCC, particles are suspended in excess solution with no capillary forces present, and after the SCC, the positions of the particles are locked. Ignorance of this locking

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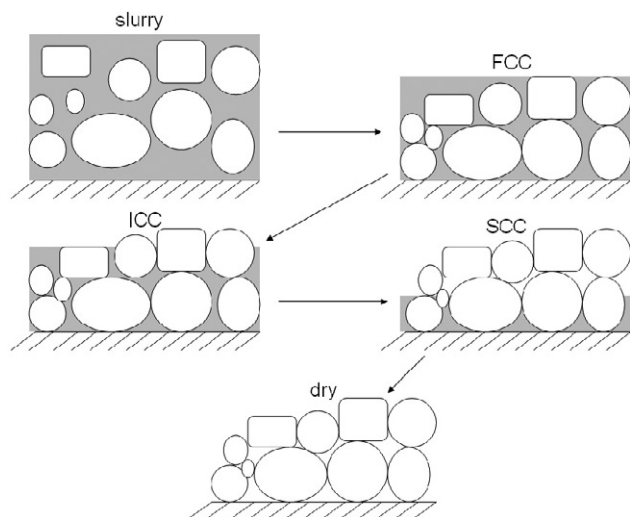


Fig. 1. Schematic representation of the drying process of a pigmented coating layer as described by Watanabe and Lepoutre [1].

effect has often led to incorrect conclusions in the literature, especially concerning so-called migration of particulate material, for example latex as reviewed by Groves et al. [2].

The experimental measurements and modelling simulations that were presented in the previous publications by the authors represent a novel approach to the study of the drying process of a pigmented coating layer. They involved measurement of the shrinkage forces [3], the study of the strength and nature of the interactions between binder and pigment [4], and the modelling of the void space and the size distributions of skeletal elements in the dry coating layer [5]. In the present study, we draw conclusions on the whole process, and postulate a model of the drying process of a pigmented paper coating layer based on a stick–slip mechanism occurring between the FCC and the SCC structures represented in Fig. 1.

2. Experimental

2.1. Materials

A ground calcium carbonate (GCC) was used as pigment and consisted of a pre-dispersed limestone from Orgon, France, ground to two particle size ranges, named Hydrocarb 60 and Hydrocarb 90 (Omya AG, Switzerland). The samples contained, respectively, 60 wt% (coarse GCC60) and 90 wt% (fine GCC90) of particles with diameter $< 2 \mu\text{m}$, and with a maximum diameter of around $5 \mu\text{m}$.

The binder for each coating formulation was either a high glass transition temperature ($T_g = 23^\circ\text{C}$) acrylic latex Acronal S320D¹ in the form of spherical particles of diameter $0.2 \mu\text{m}$, or a low T_g (5°C) styrene–butadiene latex DL930² with spherical particles of diameter $0.15 \mu\text{m}$. The minimum film-forming temperatures are given by the manufacturers as 1°C above T_g . Therefore the low T_g latex was expected to film-form, as much

as the coating structure would allow, at the temperature of the experiment ($20 \pm 1^\circ\text{C}$), whereas the high T_g latex was not.

The total solid content of the coating colours was 55 wt%, 25 wt% of which was binder. The Acronal S320D + carbonate coating formulations had pH 8.5 and conductivity 1.4 mS cm^{-1} , while the DL930 + carbonate formulations had pH 8.7 and conductivity 2.4 mS cm^{-1} . The amount of coating colour applied onto the substrates was 10 g m^{-2} , corresponding to a dry coating layer thickness of $5 \mu\text{m}$.

2.2. Shrinkage forces

A series of coating colours was prepared as described. They were applied with a laboratory coater onto strips of an elastic filled synthetic substrate (Synteape³ [3,6]), and also aluminium foil, and dried at room temperature. After being coated, the Synteape-based strips were clamped at one end and allowed to deform in the vertical plane. As deformation progressed, the extent and shape of the curl was monitored, as was the water loss. Each strip of synthetic substrate was assumed to be an elastic bending beam. Standard bending theory was applied which relates the forces acting on a beam with the actual deflection of the beam itself. The forces acting on the surface, causing the deformation, could then be calculated if the Young's modulus of the substrate is known. Four main assumptions were made: (i) the weight of the substrate was negligible when compared with the forces acting on its surface, (ii) the mechanical properties of the substrate did not change during the drying of the coating layer, (iii) the substrate acted perfectly elastically in the direction of curl, and (iv) the resistance to bending of the drying coating colour layer, which was $5 \mu\text{m}$ thick, was negligibly small when compared to that of the $100 \mu\text{m}$ -thick substrate. The theory and assumptions were supported by the observation that the samples curled into the shape of the arc of a circle (Laudone et al. [3]).

2.3. Mercury porosimetry

The dry structures of the coating layers applied onto aluminium foil were analysed by mercury intrusion porosimetry. The measurements were carried out using a Micromeritics Autopore III mercury porosimeter, able to apply a maximum pressure of mercury of 60,000 psia (414 MPa).

The results of the measurements were corrected for sample compression by applying the equation of Gane et al. [7], incorporated into the software Pore-Comp.⁴ The corrected mercury intrusion curves were used to create a network model of the void space and the structural elements [5], which is described in Section 4.

2.4. Rheology

The rheological properties of coating colour formulations have been the subject of many studies, mostly related to the

¹ Acronal is a tradename of BASF, Ludwigshafen, Germany.

² DL is a product of emulsion polymer from Dow Chemical, Midland, Michigan, USA.

³ Synteape is a product name of Arjo Wiggins.

⁴ Pore-Comp is a software package of the Environmental and Fluid Modelling Group, University of Plymouth, PL4 8AA, UK.

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