



The effect of anionic-cationic multilayering and microstructure on dye-based ink absorption

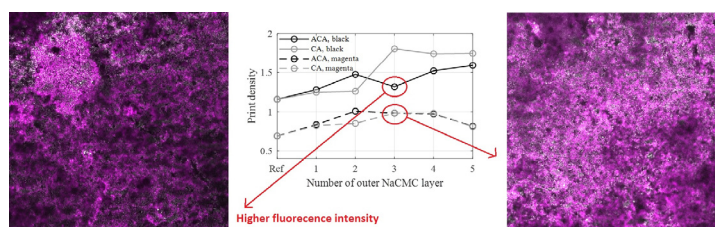
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HIGHLIGHTS

- Minerals can be more efficiently used if preparing multilayer coatings.
- Multilayering is essential for dye setting and dye capturing mechanism.
- Dye penetration and retention in the formed hydrophilic gel complex can be controlled.
- A larger amount of the binder resulted in a lower porosity of the coatings.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of water-soluble anionic binders and a modified cationic-precipitated calcium carbonate layer-by-layer structure on dye ink spreading and absorption has been studied. The multilayered coating was alternately spray-coated onto the substrate in order to determine dye interdiffusion and specific interaction including solvent absorption behavior in the nano- and microporous layers. The amount of the hydrophilic binder in the top coating was varied in order to adjust the porosity and to determine the effect of electrostatic charge interaction and to adjust dye fixation. Cationic nano- and microporous coatings and particle assemblies were created by spray-coating with cationic calcium carbonate precipitated in the presence of PDADMAC. The deposition of dye on the various coatings revealed that a small amount of anionic binder in the top coating favoured the absorption and spreading of dye and solvent, whereas a higher binder dose closed the pores and reduced the dye fixation and rate of solvent absorption. Although the top layer is essential for dye retention, it was demonstrated that the multilayering is essential not only for dye setting and dye capturing mechanism but can be utilized for vertically controlling the dye penetration and retention in the formed hydrophilic gel complex, which has a significant influence on optical effects and mechanical durability.

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1. Introduction

Liquid spreading and absorption on composites or fiber-based substrates are strongly dependent on interfacial interaction, surface roughness and surface or substrate permeability. For complex liquids such as dyes and inks, the absorption behavior is important for both durable prints and high image reproducibility but adequate ink immobilization, fixation, coalescence and drying are also important (Malla and Devisetti, 2005; Lundberg et al., 2010).

Abbreviations: CLSM, Confocal laser scanning microscope; DIM, Diiodomethane; EG, Ethylene glycol 1,2-ethanediol; GCC, Ground calcium carbonate; NaCMC, Sodium carboxymethyl cellulose; PCC, Precipitated calcium carbonate; PDADMAC, Polydiallyldimethylammonium chloride; PVOH, Polyvinyl alcohol; SEM, Scanning electron microscope; SMAI, Styrene maleic anhydride imide.

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Dye-based inks, especially anionic dye colorants, naturally interact with cationic substituents or chemicals in the coatings. Cationic additives not only improve colorant fixation and the subsequent print quality but can also be tailored to adjust the liquid uptake and durability of the coatings (Kaimouz et al., 2010). The latter is of particular importance for wetted substrates or under moist conditions. A proper colorant fixation chemistry leads to high optical density, brighter color and sharper lines, low bleeding and low print through and preferably high rub and wet resistance (Kettle et al., 2010; Jonckherree and Mabire, 2003).

In coatings designed for aqueous-based inkjet inks, multiple components and extenders are usually required to control and enhance liquid uptake, absorption capacity and lateral solvent and colorant flow. This can be done using, for instance, nanopigments (Storbeck et al., 2005; Lamminmäki et al., 2013; Jiang et al., 2014; Kumar et al., 2012; El Saeed et al., 2015), porous absorbents or engineered minerals, such as ground calcium carbonate (GCC) (Santos and Velho, 2002) or precipitated calcium carbonate (PCC) (Hladnik and Muck, 2002). The coating structure with a high concentration of fine diameter pores combined with a large pore volume (Lamminmäki et al., 2011) leads to efficient separation of the ink vehicle from the colorant (Schoelkopf et al., 2000a), and also shortens the ink drying time (Lamminmäki et al., 2012).

Donigian et al. (1998, 1999) studied the effect of PCC on the fastness of the ink and found that water fastness can be improved when PCC is used in the coatings. Glittenberg et al. (2003) used PCC together with starch as a binder to create a coating formulation that enabled to create new printing paper grade between uncoated and premium-grade paper qualities. Sreekumar et al. (2005) blended PCC together with polyvinyl alcohol PVOH and styrene maleic anhydride imide SMAI, and reported an improvement in image sharpness.

The effect of anionic binders, however, is usually controversial or lacks reliable information. Nilsson and Fogden (2008) used sodium carboxymethyl cellulose (NaCMC) as a binder in thin pigment coatings and showed that the color gamut and optical density depended on the porosity and pore size. The challenge, however, is to find an optimal nanostructure coating having a high ability to quickly absorb the carrier medium (solvent) while the colorants are preferably fixed within the coating layer to provide the best optical result and strong adhesion with the substrate. The use of larger amounts of nanoparticle binders in combinations with nano- or micro-sized pigments might lead to a liquid-repelling effect, causing intercolor bleeding and reduced dye fixation. Multilayer coatings with cationic and anionic polyelectrolyte multilayers has earlier been studied. It has been shown that adjustment of the surface chemical properties and nanoscale surface properties by deposition of small amounts of chemicals can lead to a significant improvement in ink substrate interaction and hence print quality or dye-based ink adhesion can be improved (Mielonen et al., 2014; Mielonen et al., 2016a).

The aim of the present investigation was to determine the effect of a layered coating composite on dye fixation and liquid spreading and wetting behavior. Different three layered coatings were prepared in order to adjust the anionicity, hydrophilicity and porosity. Different amounts of a hydrophilic anionic binder were used to adjust the solvent absorption rate and also the pore and cationic

site accessibility. The role of the pre-coating binder layer was also studied. The middle layer consisted of a highly cationic precipitated calcium carbonate (PCC) substantially free from binder, the availability of which was adjusted by adjusting the resin (binder) layer in the top coating. The liquid and dye absorption and spreading characteristics were assessed from contact angle measurements and drop absorption rates. The pore size distribution of the coated samples was characterized by capillary flow porometry. The print density, ink bleeding and wicking and print mottle were determined on the printed samples and confocal laser scanning microscope images were used to locate and visualize the dye spreading and absorption mechanism.

2. Materials and methods

2.1. Base papers, coating methods and chemicals

Different layered ink-receptive coating structures (Table 1) were prepared by depositing anionic (A) and cationic (C) coatings alternately onto a cellulosic substrate (290 g/m² uncoated paperboard, size A4) using an automatized and a high precision nanospray coating unit (Spalas Coating System, Nanotrons, USA).

A hydrophilic anionic sodium carboxymethyl cellulose water-soluble polymer (Finnfix[®] 5, CP Kelco, Finland) was used to spray an ink-solvent-absorbent pre-coating, and synthesized cationic precipitated calcium carbonate (PCC) particles were deposited on top of the pre-coating layer with spray coater (1st coating) in order to provide cationic sites and a specific dye fixation layer. A top coating consisting of low molecular weight NaCMC was sprayed onto the 1st coating to provide particle adhesion, and to determine the effect of pore clogging on solvent uptake and dye fixation (2nd coating). The amount of 2nd coating varied from one to five layers. A similar test series, with 1st and 2nd was made without the first pre-coating layer to clarify the effect of the first binder layer (pre-coating) on the ink absorption behavior. The targeted coat weight of the pre-coating layer and one 2nd coating layer was 1 g/m²/layer and for the 1st coating layer 3 g/m².

Cationically modified precipitated calcium carbonate was synthesized via carbonation of calcium hydroxide with CO₂ in the presence of a cationic functional chemical (polydiallyldimethylammonium chloride PDADMAC, Catiofast[®] BP, BASF). The chemical and precipitation conditions were chosen to adjust and control both PCC morphology and the surface chemistry. The ζ -potential of the PCC suspension was +31.7 mV (Malvern Zetasizer Nano ZS) after dilution in a 0.01 M NaCl solution and pH adjustment to 9.1.

The pre-coating layer and 1st coating layers were dried before the 2nd coating layer were applied according to a wet-on-wet principle without intermediate drying between each layer. NaCMC was diluted in tap water to a solids content of 5 wt-% and the solids content of PCC was 14.7 wt-%. The Brookfield viscosity (100 rpm, spindle number 5) of the NaCMC was 133 cP and of the PCC 20 cP.

2.2. Scanning electron microscope

Scanning electron microscope (SEM, Hitachi SU3500 with Hitachi SE detector) images were using 5.00 kV accelerating voltage and a 4360 μ m working distance captured to show the shape and

Table 1
The design of the coating structures.

Sample name	ACA1	ACA2	ACA3	ACA4	ACA5	CA1	CA2	CA3	CA4	CA5
Anionic layer (pre-coating)	1×	1×	1×	1×	1×					
Cationic composite layer (1st coating)	1×	1×	1×	1×	1×	1×	1×	1×	1×	1×
Anionic binder layers (2nd coating)	1×	2×	3×	4×	5×	1×	2×	3×	4×	5×

1×–5× means the number of layers.

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