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### Progress in Organic Coatings





# The influence of starch-based bio-latex on microstructure and surface properties of paper coating



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#### ABSTRACT

The microstructure of coating layers has important effects on final properties of paper. Eco-friendly bio-latex derived from sustainable starch has being increasingly used in paper coatings to substitute part of the petroleumbased synthetic latex as pigment binder. The influence of starch-based bio-latex on microstructure and surface properties of coatings such as surface composition, surface morphology, void fraction and water absorbency was studied in this paper. X-ray photoelectron spectroscopy (XPS) results showed that bio-latex addition led to a nearly linear decrease in surface carbon content for coatings dried at high temperature. Scanning electron microscope (SEM) images demonstrated that bio-latex addition may cause a lack of binder film at the coating surface that binds pigment particles, which was in agreement with the XPS results. The coatings demonstrated a marginal increase in surface roughness and some decrease in gloss with the addition of bio-latex, as was expected when starch-based binder was used. Unexpectedly, the void fraction of coatings increased slightly, which was contrary to the case in conventional cooked starch. The coatings had a significant decline in water contact angle after bio-latex addition, indicating a considerable increase in water absorbency. The changes in coating microstructure may be attributed to the unique core-shell structure and water-swollen nature of starch-based bio-latex particles.

#### 1. Introduction

Papers are often coated for aesthetic reasons to enhance surface performance and improve print quality. The quality of coated paper product is basically determined by the coating composition and microstructure, such as the types of binder and pigment used, their distribution at the coating surface and throughout the thickness, and the pore structure of coatings, etc [1–5]. It is well recognized that insufficient binder concentration in coating layer may lead to linting and dusting problems during printing process. Uneven binder enrichment at the coating surface may bring about uneven ink transfer and non-uniform water and ink absorbency, thereby causing print mottles [3,4,6–9]. Void fraction and pore size distribution are important in controlling the coating absorptivity [1,2,10]. The increase in binder content and decrease in porosity usually cause absorbency to decline. Therefore, it is important to understand the relationship between properties and microstructure of coatings.

Great attention has been put into environmental aspects in recent years, and considerable effort has been made to the replacement of fossil-based materials with environmentally friendly, natural resources derived products. Starch-based bio-latex is a newly developed crosslinked water-swollen starch nanoparticles obtained by transforming renewable crop resources into nanoparticle agglomerate. It has begun to partially replace traditional petroleum-based latex such as styrene butadiene (SB) latex, styrene-acylic (SA) latex and polyvinyl acetate (PVAc) latex in the production of coated papers [11–14]. When the substitution ratio is less than 35%-50% in the coating formulation, the final coated paper products showed good performance in properties such as gloss and surface strength, comparable to when single synthetic latex binder system is used. However, when the substitution ratio increases further, the properties of coated paper such as gloss, print surface strength and wet pick performance, would deteriorate. For the sustainable development of coated paper industry, it is therefore necessary to find new solutions to achieve higher substitution ratios.

Previous work reported in the area of starch-based bio-latex binder mainly focused on the production process and final paper properties [11–13]. No published literature concentrated on the effects of bio-latex on surface chemistry and absorbency of the coating, which can indicate the eventual print performance. The objective of this paper is to investigate the influence of starch-based bio-latex on the microstructure

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and water absorption properties of coating layers. Two types of binders, i.e., starch-based bio-latex and conventional styrene-butadiene (SB) latex, were used with kaolin clay pigment to prepare model coating layers. The total binder level in the coating formulations studied was kept constant at 5 pph (parts per hundred parts of pigment, on a dry weight basis), but the dosage ratio of two binders varied. Properties of the coating layers such as surface composition, pore structure, gloss, roughness and the resulting absorbency were examined to evaluate the influence of bio-latex on coating layer microstructure.

#### 2. Experimental

#### 2.1. Materials

The starch-based bio-latex used (EcoSphere 2330) was a commercial product of EcoSynthetix Incorporation of the USA, which had an average particle size of about  $0.10 \,\mu$ m. The SB latex (HZ 616) had an average particle size of about  $0.12 \,\mu$ m and solids content of 50.0%, and was supplied by a local latex binder manufacturer (Haoze Chemicals, Cangzhou, China). A kaolin clay of commercial coating grade with 95–96% (by weight) of particles finer than  $2 \,\mu$ m and an average particle size of about  $0.85 \,\mu$ m, supplied by Maoming Technology, China, was used as the mineral pigment. An industrial grade sodium polyacrylate (DC-40) with solids content of 42%, supplied by Wanhe Chemicals, Weifang, China, was used as pigment dispersant. Non-absorbent polyester (Mylar) film with a thickness of about 180  $\mu$ m was used as the coating substrate.

#### 2.2. Preparation of coatings

The kaolin clay pigment was pre-dispersed at 70% solids content in distilled water, using 0.3 pph of sodium polyacrylate as the dispersing agent. The starch-based bio-latex powder was pre-dispersed in distilled water at 20% solids content under mechanical agitation to form a uniform dispersion. The clay slurry, bio-latex dispersion, SB latex and additional distilled water were mixed in a mechanical mixer to prepare model paper coating colors, following pre-designed formulation recipes. The total binder level (including bio-latex and SB latex) in the coating formulations was kept constant at 5 pph, while the dosage ratio of the two binders varied. The solids content (including pigment and binders) of coating colors were kept constant at 50%. The pH of coating suspensions was adjusted to 8.0–8.5 with sodium hydroxide solution.

The coating colors prepared above were applied on the non-porous polyester film with a wire wound draw down bar to form model coating layers. The use of polyester substrate can ensure that all water lost is by evaporation from the coating surface, so that binder composition at the coating surface can be characterized free of the interference of water and binder penetration into the substrate. However, in the industrial situation, fibrous paper was usually used as the coating substrate, which may cause a little difference between the results due to the absorption of water from the coating into the base paper. The wet coating layers were dried at ambient condition (about 23 °C, 40–50% RH) or in a hot air circulation oven at 80 °C with a wind speed of about 0.2 m/s. The drying rates were slower than industrial situation. The resulting coat weight was in the order of  $30 \text{ g/m}^2$ .

#### 2.3. Characterization of coating surface binder composition

X-ray photoelectron spectroscopy (XPS, Thermofisher ESCALAB 250Xi) was used to determine the binder composition at the coating surfaces. XPS measurements were performed with a monochromatic Al K $\alpha$ X source (1486.6 eV) in a working pressure lower than 10<sup>-9</sup> mbar. Surface elemental composition (in atomic concentration percentage) was calculated from peak heights by using theoretical sensitivity factors provided for the instrument. The data reported are from average values of three samples [14].

#### 2.4. Characterization of coating surface morphology

The surface morphological characteristics of coating layers containing different amounts of bio-latex was imaged with a scanning electron microscope (SEM, Hitachi S3400). All coating samples were coated with gold before detection to increase electrical conductivity and overcome charging problem. The acceleration voltage of electron beam used was 15 kV.

The surface roughness of coatings was measured using a Parker Print Surf tester (PPS) at a clamping pressure of 1 MPa with a hard backing. Coating gloss was measured with a Hunter gloss meter at an incident angle of  $75^{\circ}$ , according to the TAPPI standard method. Five different samples were measured for each coated surface. The PPS and gloss values reported were the average values of each coating.

#### 2.5. Pore structure determination

Mercury intrusion method was used to determine the pore structure of coating layers with an automatic porosity tester (AutoPore IV 9500). It is based on Washburn equation deduced from non-wetting capillary principle [10]. Mercury content was measured under external pressure, and then converted to pore volume and pore diameter size distribution. The low pressure range was set from 0 to 30 psi, and the high pressure range was set from 30 psi to 30000 psi. Three different samples were measured for each coating, and the average value of each coating was reported.

#### 2.6. Characterization of coating absorbency

The absorbency of coating layers was characterized with a contact angle measuring system (DSA20, Kruss) at room temperature [5,15]. The shape changes of probe liquid drop on the coating surface were recorded with a microscope video system during the test period, and then analyzed with image analysis software. The circle fitting method was used to evaluate the drop shape and determine the values of contact angle. The size of sessile water drop was 10  $\mu$ l, as it can get good balance between accuracy and ease of use [15]. The average value of five measurements was reported.

#### 3. Results and discussion

#### 3.1. Coating surface binder composition

The surface carbon elemental content determined with XPS for coatings containing various proportions of bio-latex/SB latex at two different drying temperatures was shown in Fig. 1. XPS is a well-established technique for characterizing elemental compositions of materials at the surface with the measuring depth of about 5–10 nm [4,5,8,14–17]. All surface elements except hydrogen and helium can be detected. In the coating formulations of this study, the C signal mainly originates from SB latex and bio-latex binder [14,15], therefore the

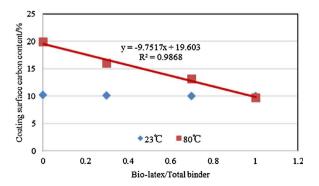


Fig. 1. Surface carbon content for coatings dried at room temperature and 80 °C.

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