The Influence of Nano-Fibrillated Cellulose as a Coating Component in Paper Coating

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This work investigates nano-fibrillated cellulose (NFC) as a component in mineral pigment paper coating. In this work, bleached *Eucalyptus* pulp was pretreated by TEMPO (2,2,6,6-tetramethyl-1-piperdinyloxy)-mediated oxidation. The oxidized pulp was then isolated to obtain NFC by sonication. Aqueous coating colors consisting of calcium carbonate, clay, carboxylated butadiene-styrene latex, additives, and NFC were prepared. The rheology of the coating colors and the surface properties of paper coated with NFC containing coating colors were determined. The rheological properties allowed NFC to be used in small amounts under laboratory conditions. Nano-fibrillated cellulose was found to improve the surface strength and smoothness of the coated paper. The water resistance of coated paper, on the other hand, decreased because of the hydrophilicity of NFC.

Keywords: NFC; Viscosity; Coating colors; Properties of coated paper

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INTRODUCTION

As an environmentally friendly bio-based material, nano-fibrillated cellulose (NFC) has gained considerable interest because of its mechanical strength, optical and electrical properties, and its biodegradability and sustainability (Hubbe *et al.* 2008; Nogi *et al.* 2009; Lavoine *et al.* 2012). For decades, NFC has been studied in paper science, architecture, electronics, cosmetics, medicine, and food science (Klemm *et al.* 2006; Nogi *et al.* 2009; Lavoine *et al.* 2012).

In paper science, studies on NFC relate to its preparation, pre-treatment, post-treatment, characterization, and rheological properties. Regarding rheological properties, researchers have found that NFC exhibits in an aqueous environment pseudoplasticity and shear thinning behavior (Herrick *et al.* 1983; Pääkkö *et al.* 2007; Missoum *et al.* 2010a; Missoum *et al.* 2010b). It is believed the pseudoplasticity and shear thinning behavior of NFC suspensions may improve the rheological properties of the coating material; as a result, the properties of coated paper may improve as well (Koepenick 2004, 2005). Researchers have used NFC as a rheology modifier additive in CMC-containing pigment coating formulations and found that NFC additives operated as a water-binding, gel-forming component (Dimic-Misic *et al.* 2013). In this work, it was also discovered that the substitution of CMC with NFC makes the introduction of new types of rheology modifiers in coating colors possible, which could lead to improved strength of coated paper. In addition to the specific rheological properties as discussed above, NFC possesses the properties of high specific area, flexibility, and crystallinity and has a high amount of hydroxyl groups. The introduction of NFC into coating material may therefore positively contribute the dry state properties and thus improve the performance of coated paper (Turbak *et al.* 1985; Koepenick *et al.* 2004; Beibei 2011).

Previous researchers have carried out experiments related to food packaging and printing (Ankerfors *et al.* 2012; Aulin *et al.* 2010a; Nygårds 2011). Researchers have reported that pure NFC coating can dramatically reduce the air permeability and oil resistance of coated paper (Syverud and Stenius 2009; Aulin *et al.* 2010b). It is now well understood that the high crystallinity of NFC combined with strong hydrogen bonding between NFCs contributes to the low porosity of pure NFC film; as a result, pure NFC coating can render paper with good oxygen and oil barrier in dry environment. As for water barrier attributes, a number of researchers modified NFC to change its hydrophilic nature to render the NFC films with water barrier (Stevanic *et al.* 2012; Österberg *et al.* 2013). Differently charged NFCs behave differently in terms of film forming. Highly charged NFCs, like TEMPO-oxidized NFCs, tend to bind more water and the film formation is faster and more stable (Eronen 2011). As for printing applications, Hamada and Bousfield (2010b) used NFC as a coating component to improve the printing quality of synthetic fiber sheets. They found that an NFC layer could greatly decrease the rate of ink absorption. Luu *et al.* (2011) treated a base paper first with alkylketene dimer (AKD) and then a coating with NFC. They also found a reduced ink penetration. Ridgway and Gane (2012) reported an improvement in surface strength and bending stiffness by incorporating the NFC in a coating layer.

Paper is coated to improve the surface properties. In this work, NFC was added to a typical coating formulation at low level. Typical coating formulation contains primarily mineral pigment. Compared to mineral pigments, NFC is very different in morphology and size. As paper coated with NFC has shown favorable oxygen barrier and oil barrier and print quality, it is interesting to study the mechanical properties, surface properties, and water resistance of paper coated with NFC as a coating component. The rheology of coating colors after the addition of NFC was determined in addition to the surface properties.

EXPERIMENTAL

Materials

For the preparation of NFC, bleached *Eucalyptus* pulp from Guangzhou paper mill (Guangzhou, China) was used as the starting material. A solution of NaClO (solid content of 10%) was supplied by Tianjin Fuyu Chemical Company (Tianjin, China), NaBr solution was acquired from Tianjin Damao Company (Tianjin, China), and 2,2,6,6-tetramethyl-1-piperdinyloxy (TEMPO) free radical was supplied by Alfa Aesar (Tianjin, China).

For the preparation of coating formulation, calcium carbonate (GCC, 90% w/w of particles $<$2 μm, median particle size 1.37 μm, ζ-potential: approximately -27.13 mV, Maoming Yinhua Corp., China), kaolin (80% w/w of particles $<$2 μm, median particle size 1.21 μm, ζ-potential: approximately -34.6 mV, Maoming Yinhua Corp., China), carboxylated butadiene-styrene latex (median particle size of about 0.158 μm, ζ-potential: approximately -35.9 mV, BASF Corp), NaOH solution (solid content of 10%), dispersant, and water-resistance agent (Polyamine Polyurea Resin) were all supplied by Guangzhou paper mill (Guangzhou, China).

Methods

NFC preparation

TEMPO-mediated oxidation was used to pretreat the wood pulp before sonication (Kitaoka *et al.* 1999). Two grams (dry weight) of wood pulp was first dispersed at 1% (w/v) concentration into 200 mL distilled water and oxidized through the addition of NaClO (5.5 mmol/g oven-dry pulp) in the presence of catalytic amounts of 2,2,6,6-tetramethyl-1-piperdinyloxy (TEMPO) (0.1 mmol/g based on oven-dry pulp) and NaBr (1 mmol/g based on oven-dry pulp). The experiment was carried out at a pH of 10 to 11 at room temperature. After 2.5 h of TEMPO-oxidation reaction, the pulp was rinsed 3 times. Then, 1 h sonication (Ultrasonic Processor, Newpower ultrasonic electronic equipments co., LTD, China) was applied to 1 wt% TEMPO-oxidized pulp (carboxylate content: 0.3 mmol/g), after which the NFC (ζ-potential: approximately -75 mv) was finally obtained. The NFC suspensions were used in the rheological tests and preparation of coating material.

Preparation of coating material

All coating colors were based on 100 parts per hundred (pph) pigments. Calcium carbonate (80 in dry parts) and kaolin (20 in dry parts) were dispersed with a high-shear dispersion mixer (Saijie Chemical Equipment CO., LTD, JFS—550, China) at 2400 rpm for 30 min initially. Then, carboxylic butadiene-styrene latex (10 pph), NFC suspensions, and water-resistance agent (0.2 pph) were mixed at 500 rpm. Next, 10% (w/w) NaOH (0.08 pph) was added to adjust the pH value to between 8.5 and 9.5. Subsequently, water was added to the mixture to reached target solids content. The ratio of NFC to pigment varied from 0 to 0.8 pph. The coating was used in the rheological tests and then was applied to the base paper (70 g/m^2, Pinna, China) via laboratory rod-coater (Zehntner GmbH, ZAA 2300, UK) at a speed of 15 mm/s, and the coating weight was set as 15 g/m^2.

Atomic force microscopy

Atomic force microscopy (AFM) images were obtained with a Nanoscope Ⅲa multimode scanning probe microscope (VEECO Company, USA). Samples were prepared by sticking the NFC films onto a mica sheet. During the experiment, the topographic mode was used. The image was scanned using tapping mode and the obtained image was then processed by flattening using Nanoscope Analysis software.

Rheological tests

Rheological measurements of coating colors with various NFC contents and suspensions were performed in a rheometer (AR550, USA) at a temperature of 25 °C. The rheological tests were carried out at different NFC content of the suspension, which were 0.8 wt%, 1.1 wt% and 1.2 wt%, and at different ratio of NFC to pigments, which were 0, 0.4 wt%, and 0.8 wt%. The configuration adopted was a cone and plate. Rheological measurements of NFC suspensions were performed with shear rates ranging from 10 to 100 s^{-1}, and the rheological measurements of coating colors were performed at shear rates that ranged from 0 to 10000 s^{-1}.

Performance tests of coated paper

Before the tests, paper sheets were held under constant temperature and humidity for 24 h. The mechanical properties, surface properties, and water resistance were then measured.

Tensile strength of paper samples was determined with a Lorentzen and Wettre tensile tester (L&W CE062, Sweden) according to ISO 1924-2 (1994).

Dry pick velocity of paper samples were tested using a standard IGT pick tester (IGT testing systems, AIC 2-5, USA) according to ISO 3783 (2006). The paper samples were brought into contact with an inking wheel, which was coated with a standard tack ink. The wheel was continuously accelerated until a portion of the paper coating had been lifted. The results of the tests were given as the printing velocity when a portion of paper was lifted. In this test, the final printing speed was set at 4 m/s.

Smoothness testing was performed using a Bekk smoothness tester (ZB-BK10, Hangzhou, China) in accordance with ISO 5627 (1995).

Whiteness of paper samples were tested in a Lorentzen and Wettre whiteness tester (Elrepho 070, Sweden) based on ISO 2470-2 (2008).

Air permeability of paper samples were tested according to ISO 5636-3 (2013) in a Lorentzen and Wettre air permeance tester (L&W 166, Sweden).

The water resistance of paper samples was evaluated with a liquid penetration dynamics analyzer according to manufacturer's instructions (Emtec Electronic GmbH, PDA.C 02, Germany). Water was chosen as the testing liquid. Testing specimens were all cut into 55×80 mm pieces and then were immersed into the testing liquid. In the testing container an ultrasonic transmitter and a receiver were equipped to record ultrasonic signals through the Z direction of paper samples during liquid immersion. The time was recorded as Max value when paper sample was completely wetted and was expressed as the time needed to wet the paper.

The water absorptiveness of paper samples was evaluated based on ISO 535 (2014) using the Cobb method. The tests were performed using Cobb tester (HK-213, Dongguan, China). The amount of water which penetrated into the paper samples during a specific time (60 s) was measured and expressed as g/m^2.

RESULTS AND DISCUSSION

Influence of NFC Addition on Viscosity of Coating Colors

The effect of NFC addition (at ratio of NFC to pigments of 0, 0.4%, and 0.8%) on the viscosity of coating colors was investigated. The solids content of the coating formulation was set at 45%.

As shown in Fig. 1, all the coating colors with different addition of NFC showed similar shear thinning behavior (Carreau and Lavoie 1993). Under conditions of low shear rates (10^{-1} to 10^{3} s^{-1}), the viscosity of coating colors increased greatly as the NFC dosage was increased. The viscosity doubled at a ratio of 0.8% compared with the coating material without NFC, showing the strong thickening effect of NFC. This phenomenon is not good for the preparation and pumping of the coating material. However, when the shear stress was high, coating colors with different contents of NFC did not show much difference. The network of coating material was destroyed under high shear stress, showing shear thinning behavior, which is favourable for the runnability of the coating machine (Triantafillopoulos 1996). Such rheological properties allow NFC to be used in small amounts under laboratory conditions. Because low NFC additional levels can increase the viscosity of coating colors greatly at low shear rates, it might not be suited for industrial use at high addition levels.

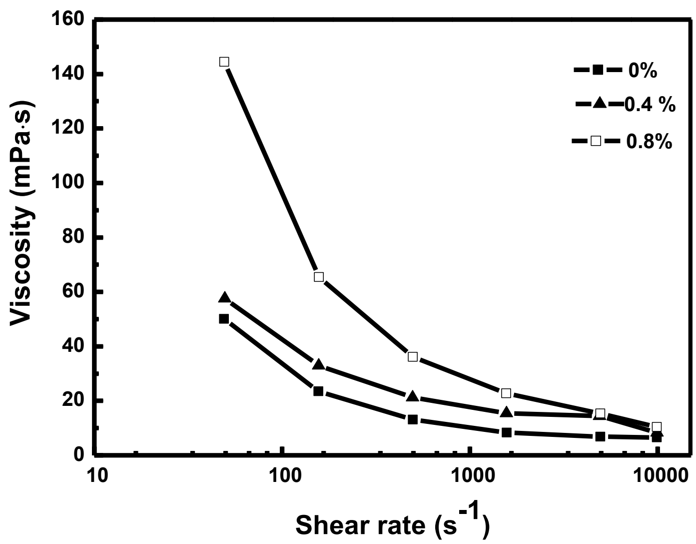

Fig. 1. The influence of NFC content on viscosity of coating colors at high shear rates

Figure 2 shows that when the NFC content was increased from 0.8% to 1.1%, the viscosity of the NFC suspension almost doubled. As shown in Fig. 3, the NFC suspension was more like gel at a dosage of 1.2%. This behavior is possible because the packing aggregation of NFC limits the formation of a continuous network, as suggested by other studies (Missoum *et al.* 2010a,b). This can also be concluded from Fig. 4, which shows the AEM image of an NFC film. The fiber diameter of NFC approximately varies between 16-32 nm while the length of individual fiber is difficult to determine because the fibers tend to cross over each other. NFC fibers aggregate firmly to form a stable network in Fig. 4. When the concentration of NFC in the suspension was increased, so was the amount of carboxylate groups, which provided repulsion within the network, thus stabilizing the network. As a result, the internal friction rose when the suspension flowed, which explains why the NFC dosage had such a marked impact on the viscosity of coating material under low shear rates. Because of the sensitivity of agglomerates to high shear stress, coating colors with higher NFC content in high shear stress did not show much difference.

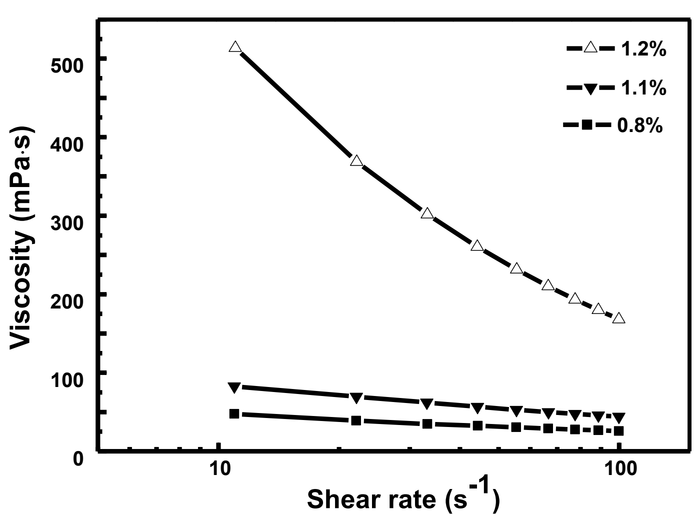

Fig. 2. The influence of NFC content on viscosity of coating colors at low shear rates

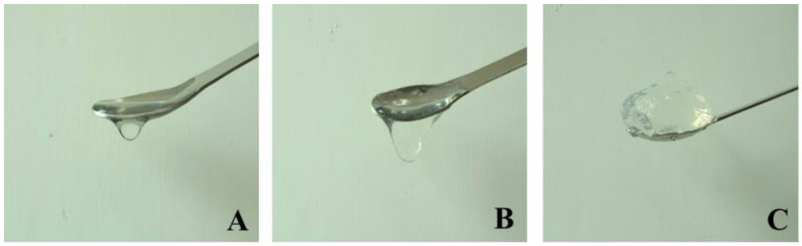

Fig. 3. Pictures of three different NFC suspensions: (A) 0.8 wt% NFC, (B) 1.2 wt% NFC, (C) 3.0 wt% NFC

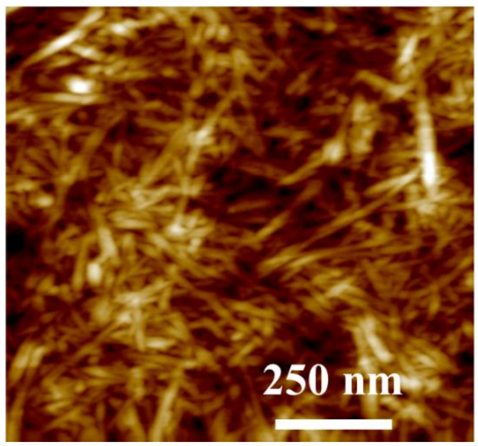

Fig. 4. AFM micrograph of NFC

Influence of the NFC-to-Pigment Ratio on Coated Paper

As stated above, the presence of NFC had a noticeable effect on the viscosity of coating colors. The rheology of coating colors also is known to influence the performance of coated paper. Therefore, the influence of NFC-to-pigment ratio on coated paper was investigated, as shown below.

The dry pick velocity of the coated paper reflects the binding strength between pigment particles as well as the coating layer and base paper. When the NFC-to-pigment ratio was increased to higher than 0.4%, the surface strength of the coated paper began to increase, as illustrated in Fig. 5. When the NFC-to-pigment ratio increased from 0 to 0.4%, the surface strength remained at the same level. However, when it was further increased to 0.6%, the effect started to show, and the surface strength of the coated paper increased by 13% compared with the 0% NFC-to-pigment ratio coated paper. When the NFC-to-pigment ratio rose to 0.8%, the surface strength of the coated paper increased by 25.5% compared with the 0% NFC-to-pigment ratio coated paper. The results showed that NFC in coating materials acted as a binder, which is consistent with the conclusions of many other researchers (Hamada *et al.* 2010a; Lavoine *et al.* 2012; Ridgeway *et al.*2012). NFC as a binder in the coating formulation could improve the binding strength between pigments and base paper.

Tensile strength is primarily used to measure the maximum load a paper sample can bear under a longitudinal loading force when it breaks. It is highly connected to the strength of fibers and the bonding strength between fibers. Consequently, the tensile strength of coated paper showed no clear effect when the NFC-to-pigment ratio increased.

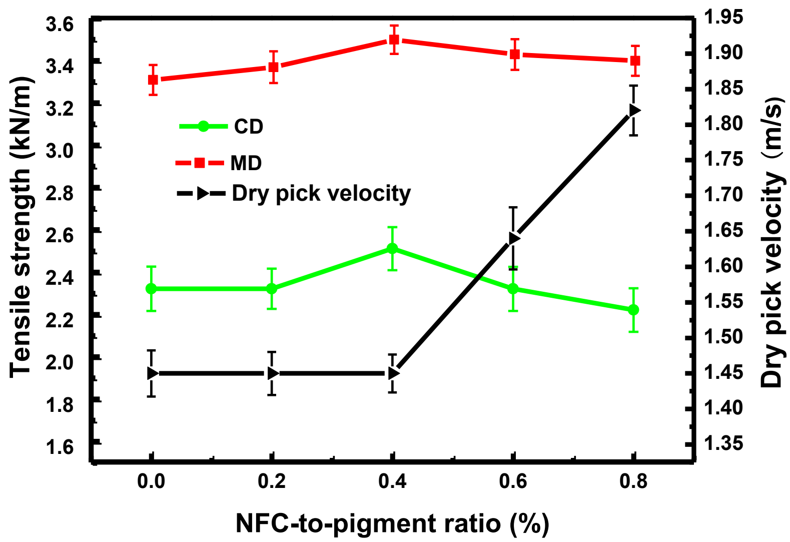

Fig. 5. The influence of NFC-to-pigment ratio on mechanical properties and dry pick velocity of coated paper
Abbreviations: Machine Direction (MD), Cross Direction (CD)

When the NFC-to-pigment ratio was 0.8%, the smoothness of the coated paper increased by 13% compared with the 0% NFC-to-pigment ratio coated paper, as shown in Fig. 6. Figure 7 shows that the air permeability of coated paper decreased as the NFC-to-pigment ratio increased. This also occurred because NFC played the role of binder in the coating material. The whiteness of coated paper showed only a slight difference when the NFC-to-pigment ratio differed because of the NFC's colorlessness.

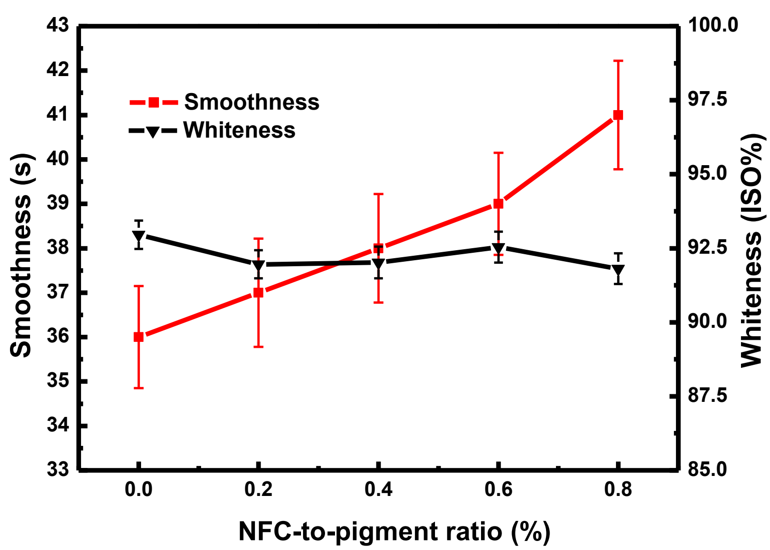

Fig. 6. The influence of NFC-to-pigment ratio on smoothness and whiteness of coated paper

It is known that paper is primarily composed of fibers that are highly hydrophilic. In addition, the porous structure of paper makes it easily permeated by liquid. In this sense, coating helps to cover the voids in paper and greatly reduces the characteristic size of pores in the paper, thus improving water resistance. However, as NFC is highly hydrophilic, the influence of NFC addition on water resistance of coated paper was evaluated. The maximum (max) value refers to the time to reach highest signal when paper sample is completely wetted in the Z direction. Higher max value indicates better water resistance of paper.

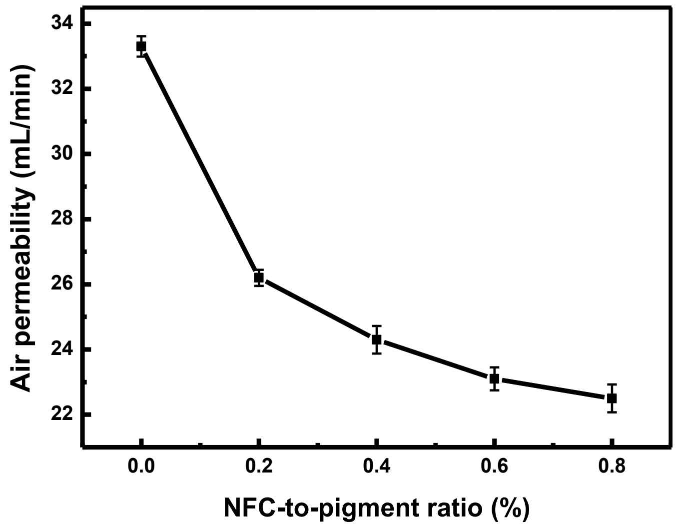

Fig. 7. The influence of NFC-to-pigment ratio on air permeability of coated paper

It can be seen from Fig. 8 that more NFC content in the coating material resulted in a lower max value of coated paper. Consequently, when the NFC-to-pigment ratio was further increased, the max value of the coated paper was reduced greatly. It could be concluded that the coated paper with NFC acting as a binder was vulnerable to water, although NFC did help improve the closure of the coating layers, thus enhancing the mechanical properties and smoothness of the paper.

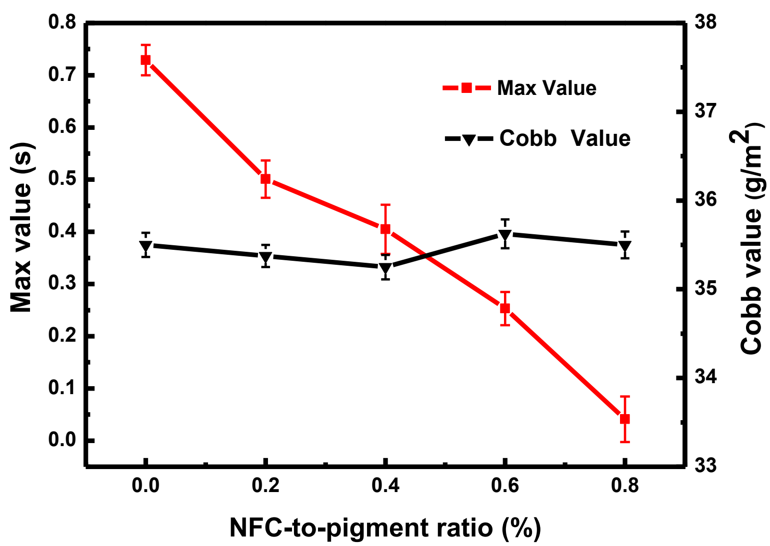

Fig. 8. The influence of NFC-to-pigment ratio on water resistance of coated paper

Cobb value is used to characterize the degree of surface sizing and internal sizing. The test determines the quantity of water that can be absorbed by the surface of paper in the given time. Based on the maximum value, the Cobb value changed little. Because the

NFC addition was small in this work, the absorbency of the coated paper did not show a large difference, which explains why the Cobb value of the coated paper changed little when the NFC-to-pigment ratio increased. However, the maximum value was measured when the paper sample was completely wetted and was expressed as the time needed to wet the paper in the Z direction. So even a little difference in the amount of NFC could be reflected in the time needed to wet the coated paper in the Z direction.

CONCLUSIONS

1. The viscosity of the coating material increased at low shear rates with increasing addition of NFC. This is possibly due to the aggregation of NFC, which limits the flow of the coating formulation. At high shear rates, no remarkable difference was observed in the viscosity of the coating material with increasing dosage of NFC, suggesting that NFC addition may not negatively affect the high-speed runnability of a coating machine.
2. Increased NFC-to-pigment ratio enhanced the surface strength of the coated paper. This suggests that NFC could be used as a binder in mineral pigment coatings.
3. Increased NFC-to-pigment ratio rendered the coated paper smoother and decreased air permeability. The application of NFC in coating is unlikely to change the whiteness of coated paper because of its colorlessness.
4. Increased NFC-to-pigment ratio lowered the water resistance of the coated paper, while the NFC addition did not affect the Cobb value of coated paper. It is because the addition of NFC was relatively small in the coating colors.

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